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In situ terminal settling velocity measurements at Stromboli volcano: Input from physical characterization of ash

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ABSTRACT

Ash particle terminal settling velocity is an important parameter to measure in order to constrain the internal dynamics and dispersion of volcanic ash plumes and clouds that emplace ash fall deposits from which source eruption conditions are often inferred. Whereas the total Particle Size Distribution (PSD) is the main parameter to constrain terminal velocities, many studies have empirically highlighted the need to consider shape descriptors such as the sphericity to refine ash settling velocity as a function of size. During radar remote sensing measurements of weak volcanic plumes erupted from Stromboli volcano in 2015, an optical disdrometer was used to measure the size and settling velocities of falling ash particles over time, while six ash fallout samples were collected at different distances from the vent. We focus on the implications of the physical parameters of ash for settling velocity measurements and modeling. Two-dimensional sizes and shapes are automatically characterized for a large number of ash particles using an optical morpho-grainsizer MORPHOLOGI G3. Manually sieved ash samples show sorted, relatively coarse PSDs spanning a few microns to 2000 μm with modal values between 180-355 μm . Although negligible in mass, a population of fine particles below 100 μm form a distinct PSD with a mode around 5-20 μm . All size distributions are offset compared to the indicated sieve limits. Accordingly, we use the diagonal of the upper mesh sizes as the upper sieve limit. Morphologically, particles show decreasing average form factors with increasing circle-equivalent diameter, the latter being equal to 0.92 times the average size between the length and intermediate axes of ash particles. Average particle densities measured by water pycnometry are $2755 \pm 50 \text{ kg m}^{-3}$ and increase slightly from 2645 to 2811 kg m^{-3} with decreasing particle size. The measured settling velocities under laboratory conditions with no wind, $< 3.6 \text{ m s}^{-1}$, are in agreement with the field velocities expected for particles with sizes $< 460 \mu\text{m}$. The Ganser (1993) empirical model for particle settling velocity

33 is the most consistent with our disdrometer settling velocity results. Converting disdrometer
34 detected size into circle equivalent diameter shows similar PSDs between disdrometer
35 measurements and G3 analyses. This validates volcanological applications of the disdrometer
36 to monitor volcanic ash sizes and settling velocities in real-time with ideal field conditions. We
37 discuss ideal conditions and the measurement limitations. In addition to providing
38 sedimentation rates in-situ, calculated reflectivities can be compared with radar reflectivity
39 measurements inside ash plumes to infer first-order ash plume concentrations. Detailed PSDs
40 and shape parameters may be used to further refine radar-derived mass loading retrievals of the
41 ash plumes.

42 **Highlights:**

- 43 • An optical disdrometer is used to measure ash sizes and settling velocities at Stromboli.
- 44 • Collected ash samples show sorted and coarse particle size distributions.
- 45 • Ash particles density and sphericity slightly decrease with augmenting size.
- 46 • Ganser's law (1993) best fits disdrometer field measurements of settling velocities.
- 47 • Volcanological applications of disdrometers to monitor ash fallout are validated.

48

49 **Keywords: Terminal Settling Velocity; Ash fallout; Particle size; Morphology;**
50 **Disdrometer; Stromboli.**

51

52 1. Introduction

53 Constraining volcanic ash plume dynamics, dispersion and fallout processes is of
54 paramount importance for the mitigation of related impacts, such as those on infrastructure,
55 transportation networks, human health (Baxter, 1999; Wilson *et al.*, 2009; Wilson *et al.*, 2012).
56 The terminal settling velocity (V_T) of particles transported in volcanic ash plumes influences
57 plume dispersal in the atmosphere, controls the sedimentation pattern in space and time, and in
58 turn, the formation of ash deposits (Beckett *et al.*, 2015; Bagheri & Bonadonna, 2016a). V_T is
59 used to estimate ash mass deposition rates (Pfeiffer *et al.*, 2005; Beckett *et al.*, 2015) and it
60 mainly depends on the total grain size distribution (TGSD), and the density and the shape of
61 ash particles. Retrieving the TGSD in real-time is currently impossible for operational purpose
62 owing to the lack of direct measurements of the *in situ* Particle Size Distribution (PSD; *e.g.*,
63 inside the plume). It is generally obtained from post-eruption analyses of ash deposits
64 (Andronico *et al.*, 2014) or from a multi-sensor strategy (Bonadonna *et al.*, 2011; Corradini *et*
65 *al.*, 2016) comprising, for instance, satellite images (Prata, 1989; Prata & Grant, 2001; Prata &
66 Bernardo, 2009) and radar remote sensing (Marzano *et al.*, 2006a, 2006b), coupled to ground
67 sampling. Meteorological optical disdrometers, although originally designed for hydrometeors,
68 can be used to record volcanic ash fallout, and provide particle number density, settling
69 velocities and sizes in near real-time at a single location. Disdrometer measurements can be
70 used to calibrate dispersion model outputs, as well as radar observations from an empirical law
71 relating derived radar reflectivity factors and associated particle mass concentrations. First-
72 order estimates of their mass loading parameters, of primary importance for hazard evaluation,
73 can then be made by comparing the calculated reflectivities to radar measurements inside ash
74 plumes (Maki *et al.*, 2016).

75 Volcanic Ash Transport and Dispersion (VATD) models require equations relating V_T
76 to particle size distribution in order to make accurate forecasts of ash dispersion and deposition.
77 As V_T also depends on particle shape parameters and densities, these need to be characterized
78 as a function of sizes. Ash particles are highly heterogeneous in shape and size due to a variety
79 of fragmentation processes (Cashman & Rust, 2016), leading to the development of empirical
80 laws describing the aerodynamic drag of the particles, from which terminal velocity depends.
81 Initially this was done for spherical grains (Gunn & Kinzer, 1947; Wilson & Huang, 1979 and
82 references therein) and then for non-spherical particle shapes based on laboratory experiments

83 (Kunii & Levenspiel, 1969; Ganser, 1993; Chien, 1994; Dellino *et al.*, 2005; Coltelli *et al.*,
84 2008; Dioguardi & Mele, 2015; Bagheri & Bonadonna, 2016b; Del Bello *et al.* 2017; Dioguardi
85 *et al.*, 2017). Such studies have revealed the need to consider the morphological aspects of ash
86 particles to refine V_T estimates, in addition to the total grain size distribution.

87 A geophysical measurement campaign at Stromboli volcano was carried out between
88 the 23rd of September and the 4th of October 2015 to characterize the mass load of ash plumes
89 and their dynamics using radars at different wavelengths, including a millimeter-wave radar for
90 ash tracking (Donnadieu *et al.*, 2016). In addition, falling ash particles were measured in-situ
91 and in real-time using an optical disdrometer and samples from ground tarps, in order to
92 constrain the PSD. The PSD is required to quantify the mass load parameters of the plume from
93 the radar reflectivity measurements.

94 In this paper, we present a physical characterization of ash particles from Strombolian
95 weak plumes using ash samples collected from ground tarps and near-ground disdrometer
96 measurements of the falling ash. **Section 2** focuses on the instruments and methodologies
97 utilized to characterize ash samples and these results are presented in **section 3**. In **section 4** we
98 present V_T measurements of ash particles obtained in the field and under laboratory conditions
99 and compare them to existing empirical models. We discuss the results and limitations and then
100 give conclusive remarks of this study in **section 5** and **6**, respectively.

101 **2. Materials and methods**

102 **2.1 Ash sampling in the field**

103 Ash samples from ash-laden plumes of Stromboli volcano were collected on the ground
104 from a 0.4 m² tarp (0.45 m × 0.9 m) and a collector (0.6 m × 0.6 m) during a Doppler radar
105 measurement campaign between the 23rd September and the 4th October 2015 (Donnadieu *et al.*
106 *et al.*, 2016). During this period, Stromboli eruptive activity was weak, producing type 2a and/or
107 2b eruptions (Patrick *et al.*, 2007), which are characterized by the emission of ash plumes rising
108 200 to 400 m high above the active vents, and drifted towards the North to the North-East with
109 prevailing winds. Six ash samples from different ash fallout events were collected on a ground
110 tarp at different locations and distances from the area of the craters: (i) two on the NE flank

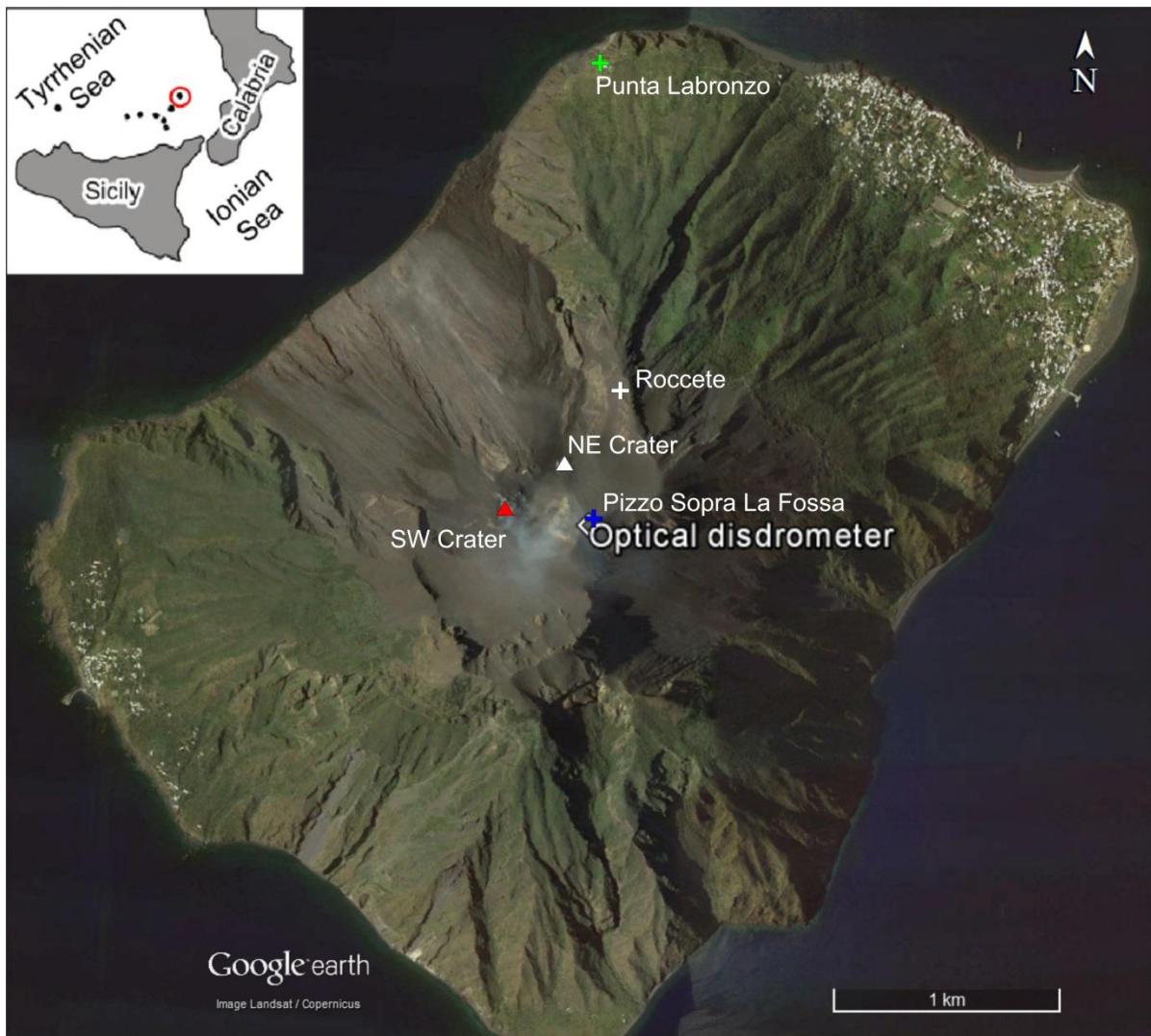
111 (Roccete) 500 to 600 m from the summit vents (white cross in **Figure 1**), (ii) three near Pizzo
 112 Sopra la Fossa, ~320-330 m northeast of the SW crater (blue cross in **Figure 1**), next to the
 113 optical disdrometer (white square in **Figure 1**), and (iii) one in a collector at Punta Labronzo
 114 ~2 km to the North (green cross in **Figure 1**). Details on ash sample collection dates and
 115 locations are summarized in **Table 1**.

116 **Table 1:** *Date and locations of the six collected ash samples.*

Date (mm/dd/yyyy)	Eruption time UTC (HH:MM)	location	Sample names	GPS point (UTM)	Collected mass (g)
09/25/2015	16:36	near Pizzo Sopra la Fossa	1636_summit	33 S 0518663 UTM 4293821	0.642
10/02/2015	12:46	NE flank (Roccete)	1246_roc	33 S 0518774 UTM 4294327	4.971
10/02/2015	15:30	Punta Labronzo	1530PL	33 S 0518720 UTM 4295743	0.259
10/02/2015	15:50	near Pizzo Sopra la Fossa	1550_summit	33 S 0518663 UTM 4293821	0.068
10/03/2015	10:42-12:52	NE flank (Roccete)	1042- 1252_roc	33 S 0518774 UTM 4294327	6.801
10/03/2015	16:01	near Pizzo Sopra la Fossa	1601_summit	33 S 0518663 UTM 4293821	25.230

117

118



119

120 **Figure 1:** Map of Stromboli Island. The Optical disdrometer was set up next to Pizzo Sopra la Fossa at
 121 900 m a.s.l. (white square), 320 m and 330 m away from the NE (white triangle) and the SW crater (red
 122 triangle), respectively. Ash samples were collected at Pizzo Sopra la Fossa next to the disdrometer (blue
 123 cross), on the NE flank (Roccete) 500-670 m NE from the vents (white cross) and at Punta Labronzo
 124 (1530PL sample, green cross) about 2 km North from the vents.

125 2.2 Grain-size and morphological analyses

126 The samples were manually sieved twice to determine their PSDs at $1/2 \Phi$ and $1/4 \Phi$
 127 intervals. The mass of each fraction was measured with a 10^{-4} g accuracy weighing scale. The
 128 relation between Φ scale and circle equivalent diameters (D) is given by $\Phi = -\log_2(D \text{ (mm)})$.
 129 In total, less than 0.5% of the mass of ash collected was lost during the $1/2 \Phi$ mechanical
 130 sieving. We calculated the sorting coefficient S_0 from Folk & Ward (1957):

$$131 S_0 = \frac{\Phi_{84} - \Phi_{16}}{4} + \frac{\Phi_{95} - \Phi_5}{6.6}, \quad (1)$$

132 with Φ_{84} , Φ_{16} , Φ_{95} and Φ_5 being the Φ values corresponding to the 84th, the 16th, the 95th and
133 the 5th percentiles, respectively, of the calculated PSD. The lower the S_0 , the more sorted the
134 PSD.

135 To study the size and shapes of ash particles, we use the MORPHOLOGI G3TM
136 automated optical analyzer (named G3 in this study) designed by Malvern InstrumentsTM.
137 Particles from a given sieve are placed on a glass plate and illuminated from below (diascopic
138 illumination). The G3's microscope measures the 2-D projected areas and shapes of a sample
139 of particles, allowing an automatic analysis of morphological parameters such as the size and
140 2-D shape parameters. We used a $\times 5$ magnification leading to an image resolution of 3.3
141 pixel/ μm^2 (*i.e.* less than 0.5 μm of minimum resolution). Typically, tens of particles of size 1
142 Φ up to 18000 particles of size $< 4 \Phi$ can be processed in 35 minutes (fast and routine analyzes,
143 Leibrandt & Le Pennec, 2015). In order to reduce the size range of the individual particles
144 analyzed while keeping them optically focused, the half- Φ fractions were sieved at a $1/4 \Phi$.
145 Obtained sieving results are presented in **Appendix A**.

146 We measure the following size parameters: (i) the longest axis (L) and (ii) intermediate
147 axis (I) in the 2-D plane orthogonal to the light direction; (iii) the circle-equivalent diameter
148 $D_{CE} = 2 \times (A_p/\pi)^{1/2}$ measured from the particle section area A_p ; and (iv) the sphere-equivalent
149 volume calculated with diameter D_{CE} . Due to the 2-D imaging inherent to the methodology, we
150 assume that particles always show the maximum projection area (Bagheri & Bonadonna, 2016)
151 and, hence, their short (S) axes are always oriented orthogonal to the image plan, *i.e.* $S \leq I$.

152 From the measurements of L , I and A_p , the following morphological parameters are
153 defined: (i) the Elongation $e = I/L$ (Bagheri & Bonadonna, 2016b); (ii) the Convexity $C_v =$
154 P_{CH}/P_p , corresponding to the textural roundness of the particles with perimeter P_p (Liu *et al.*,
155 2015), and P_{CH} being the convex hull perimeter (*i.e.* the smallest convex polygon containing all
156 pixels of the analyzed particle); (iii) the solidity $S_d = A_p/A_{CH}$ (Cioni *et al.*, 2014), indicative of
157 the high wavelength (*i.e.*, morphological) roughness of the particles (Cioni *et al.*, 2014; Liu *et*
158 *al.*, 2015b) with A_{CH} being the convex hull area; and (iv) the sphericity $\varphi = 4\pi A_p/P_p^2$ as an
159 indicator of the roughness and the shape of the particles (Riley *et al.*, 2003). The sphericity φ is
160 equal to the square of the circularity C_c (*i.e.* equal to $2(\pi A_p)^{1/2}/P_p$) defined by Leibrandt & Le
161 Pennec (2015). According to Liu *et al.* (2015a), the shape parameters associated to the convex
162 hull, such as S_d and C_v , characterize the roughness of particles independently of their form.

163 These parameters range from 0 to 1 (*e.g.*, a perfect sphere has a value of 1) and are all described
164 in Leibrandt & Le Pennec (2015), Liu *et al.* (2015a, 2015b) and Riley *et al.* (2003).

165 A complete PSD from G3 analyses, comprising all analyzed 1/4 Φ sieved fractions, is
166 estimated by combining (i) the measured mass fractions from 1/4 Φ sieving with (ii) the sphere-
167 equivalent volume (V_{SE}) of particles measured by the G3 for each analyzed fraction:

$$168 \quad m_i^{wt\%} = \sum_j \left(\frac{V_{SEi,j}}{\sum_i (V_{SEi,j})} m_j^{wt\%} \right), \quad (2)$$

169 where subscript i denotes the size bin containing individual particles analyzed by the G3 having
170 a D_{CE} diameter within the upper and lower bounds of the bin, whereas subscript j stands for the
171 sieve size fraction from manual sieving. Each bin i has a 5 μm resolution and the uncertainty
172 associated with the G3 image resolution is thus considered as negligible. $m_i^{wt\%}$ is the weight
173 fraction of particles in the i th bin size, and $m_j^{wt\%}$ is the mass percentage of the analyzed sieve
174 fraction j of the total sample mass. The ratio $V_{SEi,j} / \sum_i (V_{SEi,j})$ is the sphere-equivalent volume
175 ratio of particles belonging to the i th bin size with respect to all particles from a sieved fraction
176 j . We use the sphere-equivalent volume derived from the G3, rather than the number of
177 particles, to minimize the error propagation in the mass calculation due to the large increase in
178 particle number with decreasing size.

179 2.3 Ash density measurements

180 The average densities of ash particles of 1/4 Φ sieved fractions of the two samples with
181 the largest mass (1601_summit and 1246_roc) are measured by water pycnometry (Eychenne
182 & Le Pennec, 2012). This method allows the estimation of ash particle density by volume
183 difference between a $9.5 \times 10^{-6} \text{ m}^3$ boro-silicate pycnometer filled with distilled and degassed
184 water and then filled with water and a known mass of ash sample.

185 The density of particles is given by:

$$186 \quad \rho_i = \frac{m_i \rho_w}{m_{w1} - m_{w2}}, \quad (3)$$

187 with ρ_i the density of the i th ash size class in $\text{kg}\cdot\text{m}^{-3}$, ρ_w the density taken to be 1000 kg m^{-3} , m_i
188 the mass of ash incorporated into the pycnometer (0.4 to 2 g), m_{w1} the mass of water required
189 to fill the reference pycnometer volume and m_{w2} the mass of water required to fill the
190 pycnometer once the ash sample has been added.

191 By using water pycnometry, we measure the average particle density of a given sieved
192 fraction. For particles between $125 \mu\text{m}$ and $700 \mu\text{m}$, we assume that water surface tension can
193 be considered as sufficiently strong to avoid vesicle and asperity filling. For this hypothesis to
194 be verified, the particles are dried in an oven before being incorporated into the water (Eycheenne
195 & Le Pennec, 2012). Thus, the measured densities correspond to the apparent densities of the
196 particles, which represents their mass divided by their solid volume and the volume linked to
197 their porosity.

198 **2.4 The optical disdrometer and particle settling experiments**

199 The optical disdrometer Parsivel², designed by OTT, uses a 780 nm wavelength laser
200 beam emitted from a transmitter to a receiver, which converts the transmitted laser light into a
201 voltage signal. Described in Löffler-Mang & Jürg (2000) and Tokay *et al.* (2014), the
202 disdrometer measures the settling velocities and sizes of particles when as they pass through
203 the laser sheet. The laser obscuration time is used to estimate the settling velocities. The longer
204 a particle takes to cross the beam, the lower the settling velocity. Then, the amplitude of the
205 laser light extinction is used to calculate the size of the particles. By measuring the number of
206 falling particles and their settling velocity class values (**Appendix B**), the disdrometer
207 calculates the number density of particles crossing the beam as:

$$208 \quad N_i(D_i) = \frac{n_i}{v_i A \Delta t d D_i}, \quad (4)$$

209 with $N_i(D_i)$ the particle number density ($\text{mm}^{-1} \text{ m}^{-3}$) of the i th disdrometer size class, n_i the
210 number of detected particles with measured settling velocity v_i (m s^{-1}), A the laser sheet area
211 ($54 \times 10^{-4} \text{ m}^2$), Δt the sampling interval (10 s) and $d D_i$ the size range (mm) of the disdrometer
212 i th size class. The disdrometer measures settling velocities between 0.05 and 20.8 m s^{-1}
213 distributed among 32 classes (Classes 1 to 22 are displayed in **Appendix B**) and detects
214 particles with diameter from $250 \mu\text{m}$ to $26 \times 10^3 \mu\text{m}$.

215 We performed in-situ measurements of falling ash during the field campaign at
216 Stromboli. The disdrometer was set up about 80 cm above the ground close to Pizzo Sopra La
217 Fossa (**Figure 1**), 320-330 m northeast of the SW crater. The disdrometer recorded the ash
218 fallout events from weak Strombolian plumes that produced the two ash samples collected from
219 ground tarps next to the disdrometer (1601_summit) and lower down the NE flank (1246_roc).

220 In order to establish the ash fallout detection limits of the disdrometer and to estimate
221 the influence of the wind on particle settling velocities in the field, disdrometer retrievals are
222 tested under laboratory conditions of no horizontal nor vertical wind. Sieved ash particles from
223 the 1601_summit sample are dropped from heights between 3 m and 11 m above the
224 disdrometer laser sheet in order to verify that terminal settling velocities were reached for each
225 sieved size fraction.

226 **2.5 Terminal settling velocity models**

227 The terminal settling velocity depends on the size, the shape and the density of falling
228 particles that affect their drag forces and hence the flow regime adopted by the ambient carrier
229 fluid. For individual particle settling, V_T is defined by the following equation (Wilson & Huang,
230 1979; Woods & Bursik, 1991; Sparks *et al.*, 1997):

$$231 \quad V_T = \sqrt{D_{CE} \frac{4g(\rho - \rho_a)}{3\rho_a C_D}}, \quad (5)$$

232 where V_T is the terminal settling velocity of the particle (m s^{-1}), D_{CE} is the circle-equivalent
233 diameter corresponding to the diameter of a circle (applicable to a sphere) with area measured
234 by the G3 for each particle, g is the gravitational acceleration (m s^{-2}), ρ and ρ_a are the particle
235 and air densities, respectively, in kg m^{-3} . Here, ρ_a is equal to 1.2 kg m^{-3} at a temperature of 15
236 $^{\circ}\text{C}$ at sea level (similar to laboratory conditions) and equals to 1.12 kg m^{-3} at 900 m a.s.l. (similar
237 to field conditions). Finally, C_D is the drag coefficient and depends on the shapes of the settling
238 particles and the Reynolds number Re , which describes the flow regime in which particles fall:

$$239 \quad Re = \frac{V_T D \rho_a}{\mu}, \quad (6)$$

240 where V_T corresponds to the settling velocity of a particle within a non-moving ambient fluid
 241 (*i.e.* the air in this case) with a viscosity μ (Pa s) equals to 1.85×10^{-5} Pa s at a temperature of
 242 15 °C at sea level (similar to laboratory conditions) and equals to 1.786×10^{-5} Pa at 900 m a.s.l.
 243 (similar to field conditions).

244 To verify that ash particles were falling at their V_T , we compared the disdrometer
 245 measurements with empirical V_T laws that are based on the following assumptions.

246 First, for particle Reynolds Number between 0.4 and 500 at an altitude of 5 km above
 247 sea level (a.s.l.) and assuming spherical particle diameters less than 1500 μm , V_T of a particle
 248 can be expressed as (Kunii & Levenspiel, 1969; Bonadonna *et al.*, 1998; Coltelli *et al.*, 2008):

$$249 \quad V_T = D \left(\frac{4g^2 \rho^2}{255 \mu \rho_a} \right)^{\frac{1}{3}}, \quad (7)$$

250 Then, we compare our results with the models of Ganser (1993) and Bagheri &
 251 Bonadonna (2016b) based on **Equation 5**, which account for non-spherical particle shapes. In
 252 such a case, the drag equations are derived from empirical analyses of particle settling velocities
 253 and are not related to the same particle shape parameters.

254 In Ganser (1993), C_D is determined as follows:

$$255 \quad C_D = \frac{24}{Re K_1 K_2} \left[1 + 0.1118 (Re K_1 K_2)^{0.6567} \right] + \left(\frac{0.4305}{1 + \frac{3305}{Re K_1 K_2}} \right). \quad (8)$$

256 K_1 and K_2 being the Stokes' shape factor and the Newton's shape factor, respectively:

$$257 \quad K_1 = \left(\frac{1}{3} + \frac{2}{3} \varphi^{-\frac{1}{2}} \right)^{-1}, \quad (9)$$

$$258 \quad K_2 = 10^{1.8148(-\log \varphi)^{0.5743}}, \quad (10)$$

259 where φ is the G3-derived sphericity (Riley *et al.*, 2003) of particles considered as isometric (I
 260 = S) and is the best shape parameter to be used in the Ganser model (Alfano *et al.*, 2011).

261 In Bagheri & Bonadonna (2016b), C_D is calculated as:

$$262 \quad C_D = \frac{24k_S}{Re} \left[1 + 0.125 \left(Re \frac{k_N}{k_S} \right)^{\frac{2}{3}} \right] + \left[\frac{0.46}{1 + 5330 \left(Re \frac{k_N}{k_S} \right)} \right], \quad (11)$$

263 with k_S and k_N , being shape factors equal to:

$$264 \quad k_S = (F_S^{1/3} + F_S^{-1/3})/2, \quad (12)$$

265 where $F_S = f e^{1.3} \left(\frac{D_{CE}^3}{L I S} \right)$ and $F_N = f^2 e \left(\frac{D_{CE}^3}{L I S} \right)$

266 and

$$267 \quad k_{N=10} = \alpha_2 \left[-\log(F_N) \right]^{\beta_2}, \quad (13)$$

268 where $\alpha_2 = 0.45 + 10 / \exp(2.5 \log(\rho / \rho_a) + 30)$ and $\beta_2 = 1 - 37 / \exp(3 \log(\rho / \rho_a) + 100)$

269 These shape factors depend on 3-D ash particle axes such as L , I , S and also the
270 elongation (I/L) and flatness (S/I).

271 Because C_D , Re and V_T are dependent on each other, we use an iterative approach to
272 determine the settling velocities with both aforementioned models. We initialize V_T using the
273 Stokes law, where $V_T^{Stokes} = (g D_{CE}^2 (\rho - \rho_a)) / 18\mu$, and then iteratively calculate Re , C_D , and V_T
274 (**Equation 5**). The iterations are stopped when the velocity difference is less than 10^{-8} .

275 Finally, we calculate V_T using the Dellino *et al.* (2005) relationship:

$$276 \quad V_T = \frac{1.2065 \mu \left(D_{CE}^3 g (\rho - \rho_a) \rho \psi^{1.6} / \mu^2 \right)}{D_{CE} \rho}, \quad (14)$$

277 where ψ is a shape factor defined as the ratio between the particle sphericity (Riley *et al.*, 2003)
278 and $1/C_c$. Thus, the combination of **Equation 5** and the drag coefficient of spherical particles
279 leads to the equation of Dellino *et al.* (2005), which does not depend on C_D and Re . **Equation**
280 **14** is only valid for Reynolds number > 60 -100 (Dioguardi *et al.*, 2018).

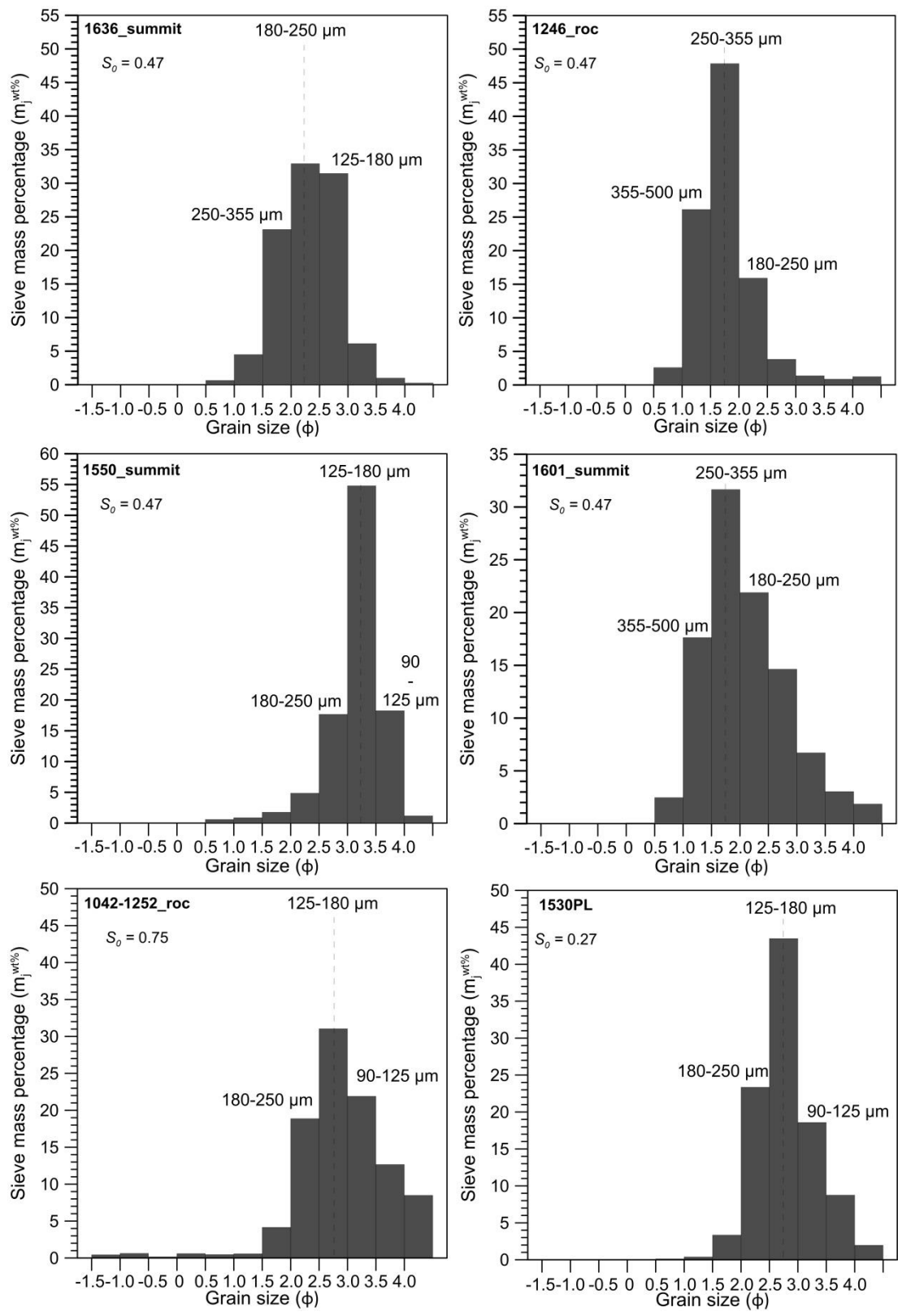
281 Ash morphological parameters are required in order to compare models of V_T for non-
282 spherical particles to ash settling velocities measured in the field or under laboratory conditions.
283 These parameters are characterized in the following section for Strombolian ash.

284 3 Ash characteristics

285 3.1 Particle size distribution by mechanical sieving and morpho-grainsizer

286 Here we present sieving results obtained for the six ash samples. Values are available in
287 **Appendix A**. The PSD from $1/2 \Phi$ sieving for the proximal samples collected on the summit
288 have modal values ranging from 125-180 μm (1550_summit) to 250-355 μm (1601_summit)
289 (**Figure 2**). The same range of PSD modes is observed for the proximal samples collected lower
290 down on the East flank at Roccete (1042-1252_roc and 1246_roc). The 1530PL sample
291 collected 2 km to the North of the summit vent at Punta Labronzo shows a mode at 125-180
292 μm . Particle sizes range from $< 63 \mu\text{m}$ to 1400 μm in 1246_roc and from $< 63 \mu\text{m}$ to 2000 μm
293 in 1601_summit (**Figure 2**). Therefore, there is no obvious correlation between sample location
294 and PSD, an observation also made by Lautze *et al.* (2013) on ash samples from type 2 eruptions
295 at Stromboli in 2009. Sorting coefficients S_0 of 0.27-0.47 indicate sorted PSDs for all ash
296 samples from a single ash plume (**Figure 2**). The higher sorting coefficient S_0 of 0.75 for the
297 1042-1252_roc sample is due to the collection of a 2-hour long succession of fallout events
298 with potentially variable PSDs, the sum of which leads to a less sorted PSD. We cannot exclude
299 some dust contamination from this sample.

300 Following the $1/4 \Phi$ sieving, the particle number frequency histogram of each sieved
301 fraction is calculated from the G3 analyses. As observed by Leibrandt & Le Pennec, 2015,
302 PSDs from the sieve fractions show a large offset toward D_{CE} values larger than the sieve mesh
303 sizes. In particular, the modal D_{CE} can lay well beyond the sieve mesh limits as shown for the
304 1601_summit sample (fractions $< 63 \mu\text{m}$ to 425-500 μm ; **Figure 3**). For example, the 250-300
305 μm sieve fraction (red PSD in **Figure 3**) actually ranges between 248-551.78 μm in D_{CE} , with
306 a mode at 350 μm , leading to 95.1% of the PSD lying above the upper sieve limit.



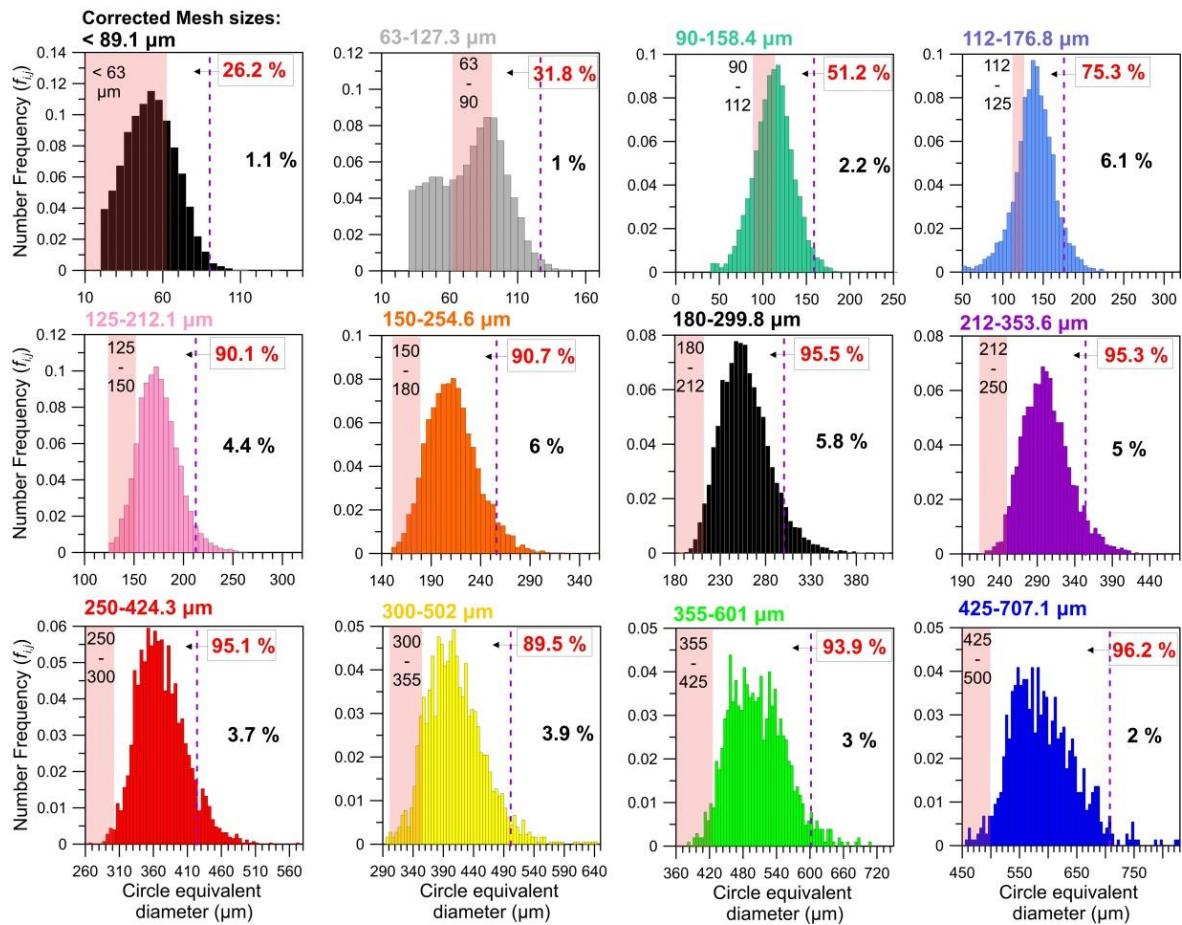
307

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Figure 2: 1/2 Φ Particle size distributions determined by manual sieving for the six ash samples of Table 1. S_0 is the Folk & Ward (1957) sorting coefficient. Lower S_0 indicates better sorting.

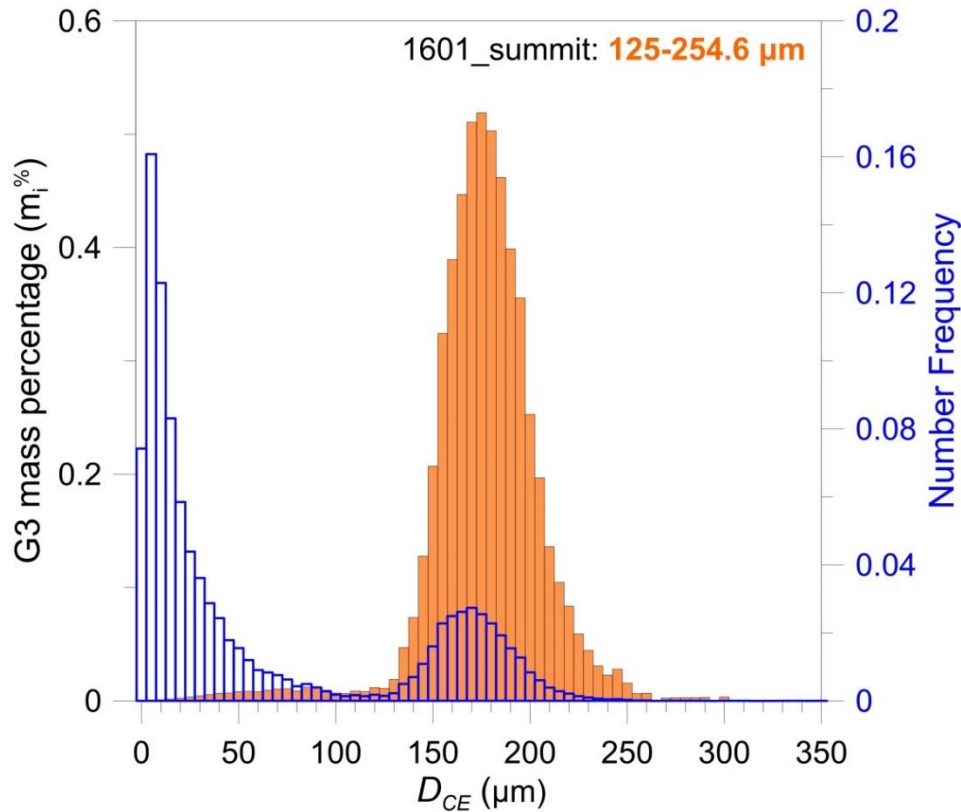


311

312 **Figure 3:** Individual particle number frequency histograms retrieved from the G3 analyses of 1/4
 313 Φ sieved fractions from the 1601_summit ash sample. Shaded red areas highlight the sieve intervals in
 314 circle-equivalent diameter and the percentage of the PSD larger than the upper sieve mesh is displayed
 315 in red. The vertical purple dashed lines indicate the diagonal dimension of the sieve upper mesh size, a
 316 better fit to the true PSD upper bound, as shown by the small residual percentage of the PSD (in black)
 317 larger than the diagonal of the sieve upper mesh. The intervals of the corrected sieve mesh sizes (down
 318 mesh size to diagonal of the upper mesh size) are indicated in color above each histogram.

319 The number proportion of the PSD lying above the upper sieve limit increases with sieve
 320 mesh size, with a minimum value of 26.2% for the < 63 μm fraction and up to 96.2% for the
 321 425-500 μm fraction. This discrepancy is explained by the fact that sieve mesh sizes (side
 322 dimension of the squared mesh) are given for supposedly spherical particles, whereas ash
 323 particles are non-spherical and often depart significantly from a spherical shape. Therefore,
 324 many particles with their largest and intermediate axes higher than the mesh size can be found
 325 in the sieved fraction depending on their orientation while passing through the mesh. The length
 326 of the squared mesh diagonal, as opposed to the mesh size, represents the true sieve upper limit
 327 when dealing with non-spherical particles, as shown by the small residual percentages of the
 328 PSD (1-6%) above the upper mesh diagonal length. Consequently, we use the lower mesh size
 329 and the diagonal length of the upper mesh, *i.e.* the upper mesh side length multiplied by $\sqrt{2}$

330 (vertical purple dashed line in **Figure 3**), to characterize the circle-equivalent diameter
 331 distributions of ash particles. These bounds of D_{CE} contain more than 94% of the ash particles
 332 in each fraction and are thus representative. In this new reference frame, for example, the 250-
 333 300 μm sieve fraction (*i.e.* $\Phi=2$) has D_{CE} lower and upper limits of 250 and 424.3 μm .



334
 335 **Figure 4:** Comparison of mass- and number-based PSD in the 125-254.6 μm sieve fraction from the
 336 1601_summit ash sample. The right axis represents the number frequency measured by G3 (blue
 337 histogram). The left axis represents the particle mass percentage $m_i\%$ (orange bars) using sphere-
 338 equivalent volumes measured by the G3.

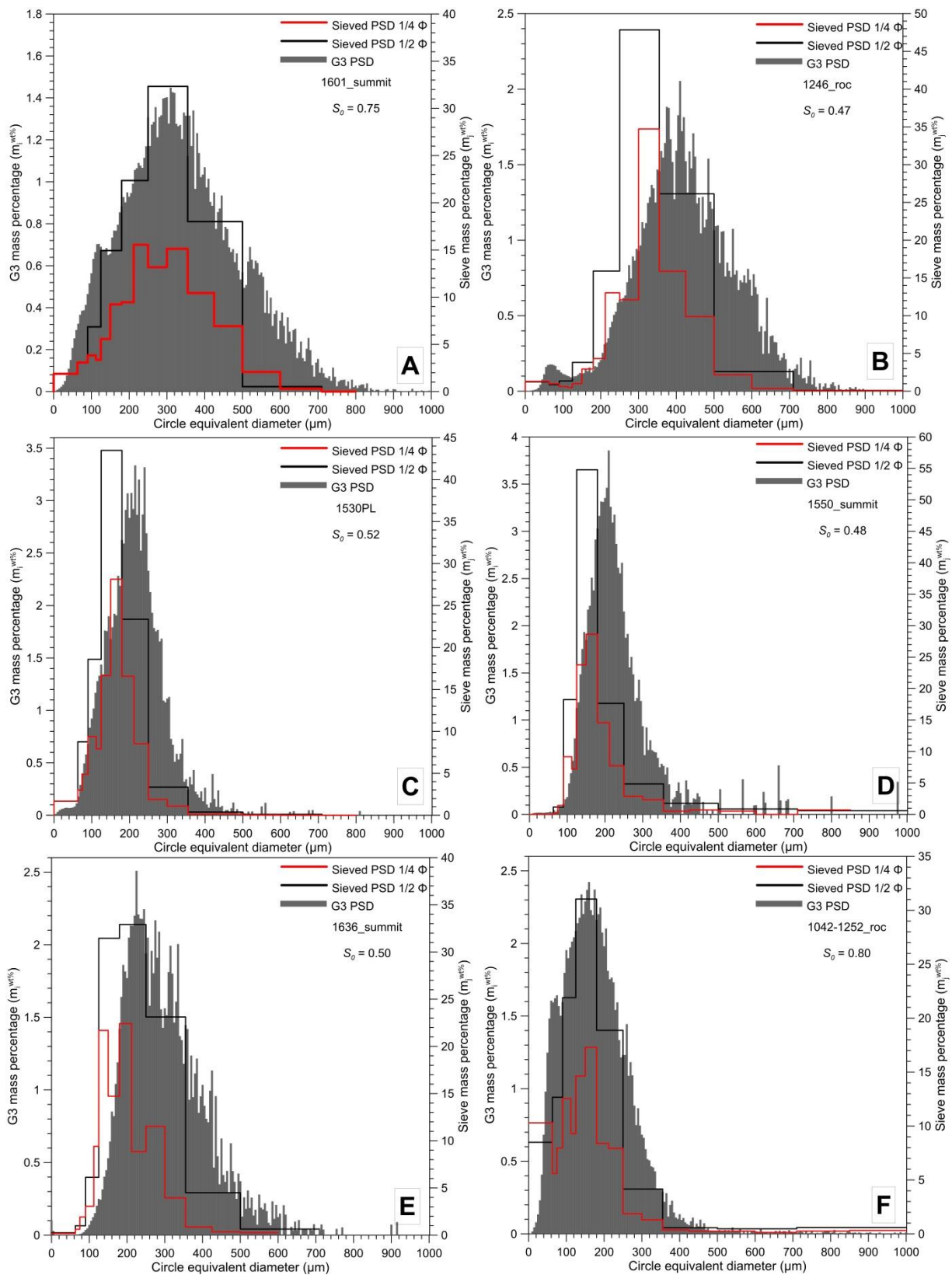
339 Every sieved fraction of the six ash samples, when analyzed in number frequency, shows
 340 a distinct population of very fine ash particles with a relatively constant modal value between
 341 5-20 μm . For example, in the 125-254.6 μm fraction of the 1601_summit sample (blue
 342 histogram in **Figure 4**), the secondary PSD of fine ash represents 73% of the total sieved
 343 fraction PSD in terms of particle number frequency (blue histogram in **Figure 4**), whereas these
 344 very fine particles represent only 3% of the whole PSD in mass or volume percentage. Likewise,
 345 in the other sieve fractions, the population of very fine ash appears as a decoupled PSD with a
 346 high contribution to the particle number frequency, but negligible in terms of mass or volume.

347 Finally, because D_{CE} distributions among successive sieve fractions exhibit a dramatic
348 overlap (**Figure 3**), we calculate mass percentages (**Equation 2**) over the whole PSD in 5 μm
349 bins by weighting the high resolution sphere-equivalent volume from the G3 analyses with the
350 mass percentage of each sieved fraction at $1/4 \Phi$. This calculation leads to high resolution (5
351 μm) mass percentage PSDs for the six ash samples (**Figure 5**). They show a unimodal
352 distribution whereas the $1/4 \Phi$ sieves display two close maxima, the latter due to splitting of a
353 unique mode at the bin transition (250 μm) into adjacent bins in **Figures 5A** and **5F**. For the
354 1601_summit and the 1042-1252_roc samples (**Figure 5A** and **Figure 5F**), the PSD obtained
355 from the $1/2 \Phi$ sieving broadly matches the corrected high resolution PSD in terms of modal
356 value, whereas the $1/4 \Phi$ PSDs tend to show a mode lower than that of the corrected PSD.
357 Unlike the 1601_summit and 1042-1252_roc calculated PSD, the other calculated PSDs are
358 well sorted (0.47-0.52).

359 The aforementioned artificial offset of the sieving PSDs toward smaller D_{CE} is more
360 obvious in the other ash samples (**Figures 5B, 5C, 5D** and **5E**), emphasizing the significant
361 bias on resulting PSD introduced by sieving non-spherical particles. Indeed, whereas spherical
362 particles would be blocked by a sieve squared mesh having its side length corresponding to
363 their diameter, coarser particles with some degree of elongation can cross the squared mesh
364 along its diagonal (side length times $\sqrt{2}$) and appear in lower (smaller mesh-sized) sieves. As
365 sieve mesh size intervals increase with diameter (*i.e.* decreasing Φ), the shift in diameter
366 increases for coarser particles. Therefore, the sieving-derived PSDs agree more closely with
367 high resolution PSDs derived from optical measurements for finer particles.

368

369



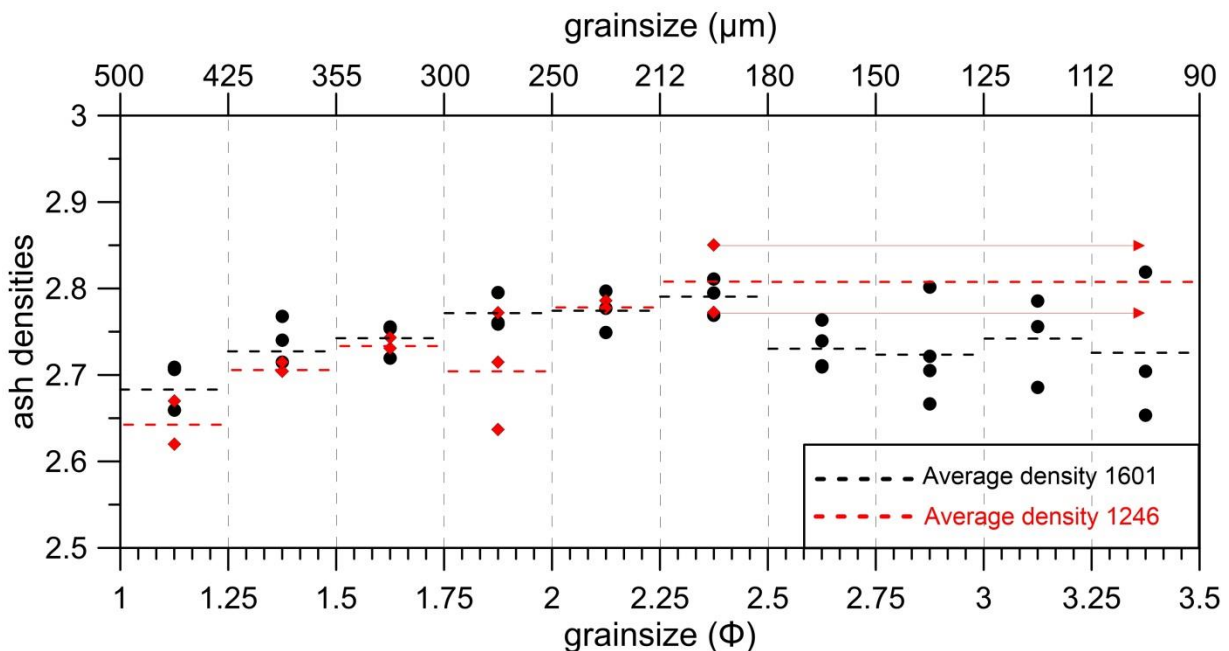
370

371 **Figure 5:** Comparison of the PSDs calculated from G3 analyses with the PSDs inferred from manual
 372 sieving (1/4 Φ, red step line; 1/2 Φ, black step line) for the six ash samples 1601_summit (A), 1246_roc
 373 (B), 1530PL (C), 1550_summit (D), 1636_summit (E), and 1042-1252_roc (F).

374

375 **3.2 Ash densities**

376 To constrain V_T , we use water pycnometry to measure the density of ash samples from
 377 the fallout detected by the optical disdrometer. Samples 1246_roc and 1601_summit have
 378 similar density trends (**Figure 6**). The average particle density of all measurements from the
 379 two summit samples is equal to $2755 \pm 58 \text{ kg m}^{-3}$. The density trend beyond $\Phi \geq 2.5$ is uncertain
 380 because the measurement's accuracy is lower for fine particles and small sample mass, as seen
 381 from the increased spread of the 1601_summit measurements for $\Phi \geq 2.5$. For this reason, we
 382 mixed the 3.5 to 2.5 Φ fractions of the 1246_roc sample to calculate a more representative
 383 average density of $2811 \pm 55 \text{ kg m}^{-3}$. In both samples, average densities slightly decrease with
 384 increasing diameter from $\Phi=2.5$ to $\Phi=1$ (*i.e.* 180-500 μm) from a maximum value of $2811 \pm$
 385 55 kg m^{-3} to $2645 \pm 35 \text{ kg m}^{-3}$. Over a particle size distribution, tephra densities typically form
 386 a sigmoidal trend that was previously described for andesitic, dacitic and rhyolitic ash
 387 (Eychenne & Le Pennec, 2012; Cashman & Rust, 2016). This sigmoidal trend is apparent for
 388 $\Phi \leq 0.5$ (*i.e.* $D_{CE} \geq 710 \mu\text{m}$) and the slight density decrease with increasing diameter might
 389 represent the beginning of a sigmoidal trend in density variation.



390 **Figure 6:** Ash densities determined by water pycnometry for the two samples 1601_summit (grey dots)
 391 and 1246_roc (red diamonds) as a function of the $1/4 \Phi$ fractions. Dashed lines correspond to the
 392 average density of each grain size class.
 393

395 3.3 Particle shapes

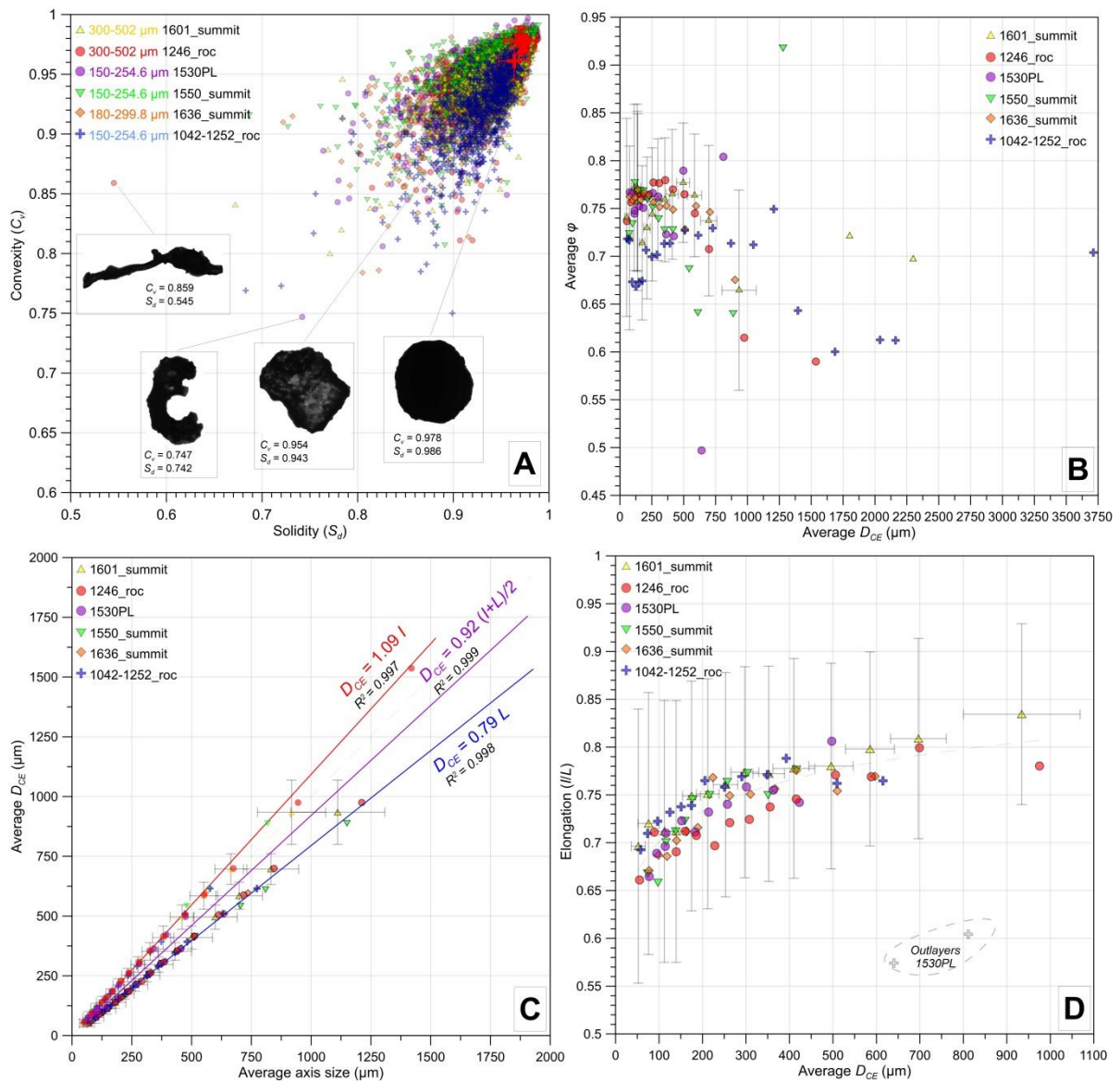
396 A comparison between S_d and C_v of the modal PSD classes of the six distinct ash
 397 samples (**Figures 2** and **5**) shows homogeneous average distributions of textural and
 398 morphological roughness among all samples (**Figure 7A** and **Table 2**). This observation is
 399 similar to morphometric analyses done by Lautze *et al.* (2011, 2013) at Stromboli showing no
 400 obvious relationship between particle shapes and the relatively short distance travelled from the
 401 source vent. In all samples, the average values of S_d and C_v are similar (**Table 2**) except for the
 402 1042-1252_roc sample, which records several fallout events over a longer collection time, as
 403 opposed to the other samples, and is possibly contaminated by wind-drifted dust. Particles show
 404 high average solidity and convexity of 0.954 and 0.943, respectively (snapshot in **Figure 7A**).

405 Though rare, irregular shaped particles can be found in several samples. Such particles,
 406 characterized by the lowest values of C_v and S_d (*i.e.* 0.747 in sample 1530PL and 0.545 in
 407 sample 1246_roc), are displayed in **Figure 7A**. In total, and among all the samples, more than
 408 90% of the analyzed particles show C_v and S_d values higher than 0.9, which characterize dense
 409 ash fragments (Liu *et al.*, 2015b).

410 Among the 6 samples, there is no clear systematic trend in sphericity as a function of
 411 particle size (**Figure 7B**). Sieved fraction average ϕ are within a narrow range between 0.7 and
 412 0.92 (< 63 to 750 μm fractions) and decrease under 0.7 to minimum values of 0.5 in the 1530PL
 413 sample. Nevertheless, with respect to the standard deviation of sphericity in the 1601_summit
 414 sample, there is no significant variation of ϕ with D_{CE} up to 750 μm , beyond which values
 415 decrease slightly.

416 **Table 2:** Average Convexity C_v , Solidity S_d and Sphericity ϕ values of the modal sieved fractions ($1/4$
 417 Φ) of the PSD for the six ash fallout samples.

Sample names	Corrected Mesh (μm)	mean C_v	Standard deviation	Mean S_d	Standard deviation	mean ϕ	Standard deviation
1246_roc	300-502	0.956	± 0.020	0.947	± 0.027	0.770	± 0.067
1601_summit	300-502	0.945	± 0.020	0.947	± 0.026	0.765	± 0.068
1530PL	150-254.6	0.955	± 0.022	0.943	± 0.031	0.762	± 0.076
1550_summit	150-254.6	0.957	± 0.023	0.939	± 0.034	0.759	± 0.081
1636_summit	180-299.8	0.955	± 0.024	0.938	± 0.034	0.757	± 0.081
1042-1252_roc	150-254.6	0.921	± 0.032	0.927	± 0.033	0.707	± 0.081



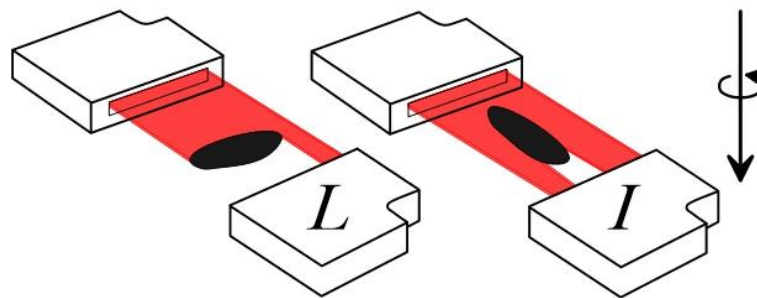
418

419 **Figure 7:** A) Clustergram of Convexity (C_v) as a function of the Solidity (S_d) of particles pertaining to
 420 the modal sieved fraction of the PSD for the 6 ash fallout samples. Average values of each population
 421 and their standard deviations are indicated in red bars and symbols. B) Average Sphericity (ϕ) of the
 422 sieved fractions as a function of their average circle-equivalent diameter (D_{CE}). C) Average Circle-
 423 equivalent diameter (D_{CE}) as a function of the average Length (L) and average Width (I). D) Average
 424 elongation (I/L) as a function of sieved fractions average D_{CE} . Errors bars (standard deviation) are
 425 shown in grey for the 1601_summit average values.

426

The observation of particle shapes is essential to understand the measurements of the
 427 optical disdrometer in terms of V_T and sizes. Here, we assume that detected ash particles tend
 428 to fall perpendicularly to the plane defined by their maximum (L) and intermediate (I) axes
 429 (Bagheri & Bonadonna, 2016). Therefore, the disdrometer should measure sizes ranging
 430 between L and I , and an average value statistically approaching $(I+L)/2$ if a random orientation
 431 of the particle L (or I) axis in the beam plane is assumed (see **Figure 8**). Taking into account

432 the linear relationships of axes dimensions with particle D_{CE} found in **Figure IV.7C** for all
433 analyzed ash particles at Stromboli, D_{CE} can be equated on average to $0.92 (I+L)/2$ with a high
434 correlation ($R^2 = 0.999$). This relationship is used thereafter to find the circle-equivalent
435 dimension of the disdrometer sized classes recording non-spherical ash particles. Hence, the
436 lower detection limit of the disdrometer of $250 \mu\text{m}$ corresponds to $230 \mu\text{m}$ in circle-equivalent
437 diameter.



438

439 **Figure 8:** Schematic representation of particle orientation when crossing the disdrometer laser
440 beam. Assuming random rotating motion and no tumbling, particles may present a length,
441 which is assumed to be equal to $(I+L)/2$.

442 With increasing D_{CE} , the I/L ratio (*i.e.* the particle elongation of Bagheri & Bonadonna,
443 2016b) increases non-linearly from 0.66 for $D_{CE} < 63 \mu\text{m}$ to 0.83 for $D_{CE} > 710\text{-}1414 \mu\text{m}$
444 (**Figure 7D**). Particles tend to be more elongated with decreasing D_{CE} . This result supports the
445 idea of an increasing proportion of particles passing through smaller sieves during manual
446 sieving, as already suggested by **Figures 3** and **5**.

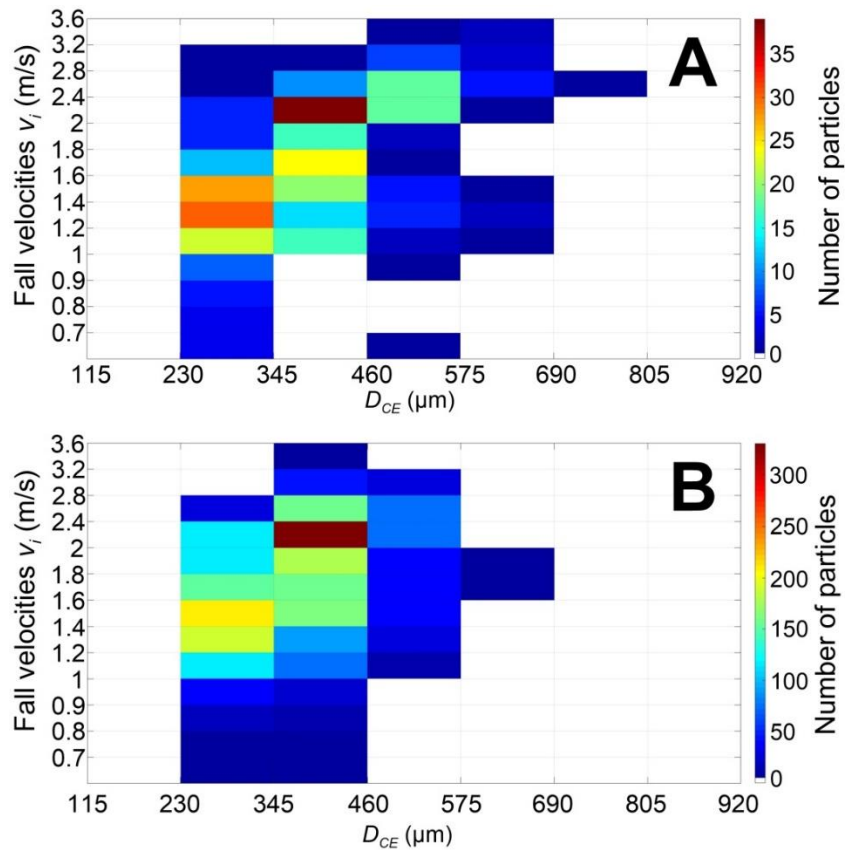
447 **Figures 7A, 7B, 7C, 7D** and **Table 2** show the overall morphological similarity among
448 all the ash samples (*i.e.* overlap in morphological parameter space) and the consistent variation
449 of the morphological parameters as a function of size. However, there is an intrinsic
450 heterogeneity existing inside each sample and each sieved fraction is characterized by: (i) the
451 individual scattering of average values of C_v as a function of S_d (**Figure 7A**) and (ii) the
452 increased spread of all shape parameter standard deviations (**Figures 7B, 7C** and **7D**). This
453 needs to be considered when interpreting disdrometer field measurements of falling ash.

454 In the next section, we use the G3's capability to measure individual particle shape
455 parameters, in order to compare V_T measured in the field by the disdrometer, as a function of
456 particle size, with existing V_T models.

457 **4. Terminal settling velocities**

458 **4.1 Field measurements**

459 The disdrometer recorded two ash fallout events on October 2 at 12:46 UTC (**Figure**
460 **9A**) and October 3 at 16:01 UTC (**Figure 9B**) totaling 355 and 2684 detected particles,
461 respectively, which were also sampled from ground tarps (1246_roc and 1601_summit
462 samples). Ash particles are detected in the first five size classes (*i.e.* $230 < D_{CE} < 804 \mu\text{m}$) and
463 the maximum number of particles (*i.e.* the mode of the PSD) occurs in the 345-460 μm class.
464 Settling velocities ranges from 0.6 to 3.6 m s^{-1} and tends to increase with particle size, as tracked
465 from their modal value across the size classes. For both events, modal V_T are comparable:
466 particles of 230-345 μm show $1.2 < V_T < 1.6 \text{ m s}^{-1}$ and those of 345-460 μm (PSD mode) show
467 $2 < V_T < 2.4 \text{ m s}^{-1}$. Particles bigger than 574 μm show $V_T \leq 3.6 \text{ m s}^{-1}$ in **Figure 9A** and $1.6 < V_T$
468 $< 2 \text{ m s}^{-1}$ in **Figure 9B**, and are present in a small amount (see PSD values in **Figures 2** and **5**).
469 Despite its lower detection limit of 230 μm (in D_{CE}), the disdrometer was able to detect at least
470 75% and 94% (in vol. %) of the particles present in the 1601_summit and 1246_roc samples
471 analyzed by the G3. In every size class, the spread of V_T around the modal value is remarkably
472 wide. In the next two sections, we focus on results of V_T obtained with a representative sample
473 with the highest collected mass (1601_summit).



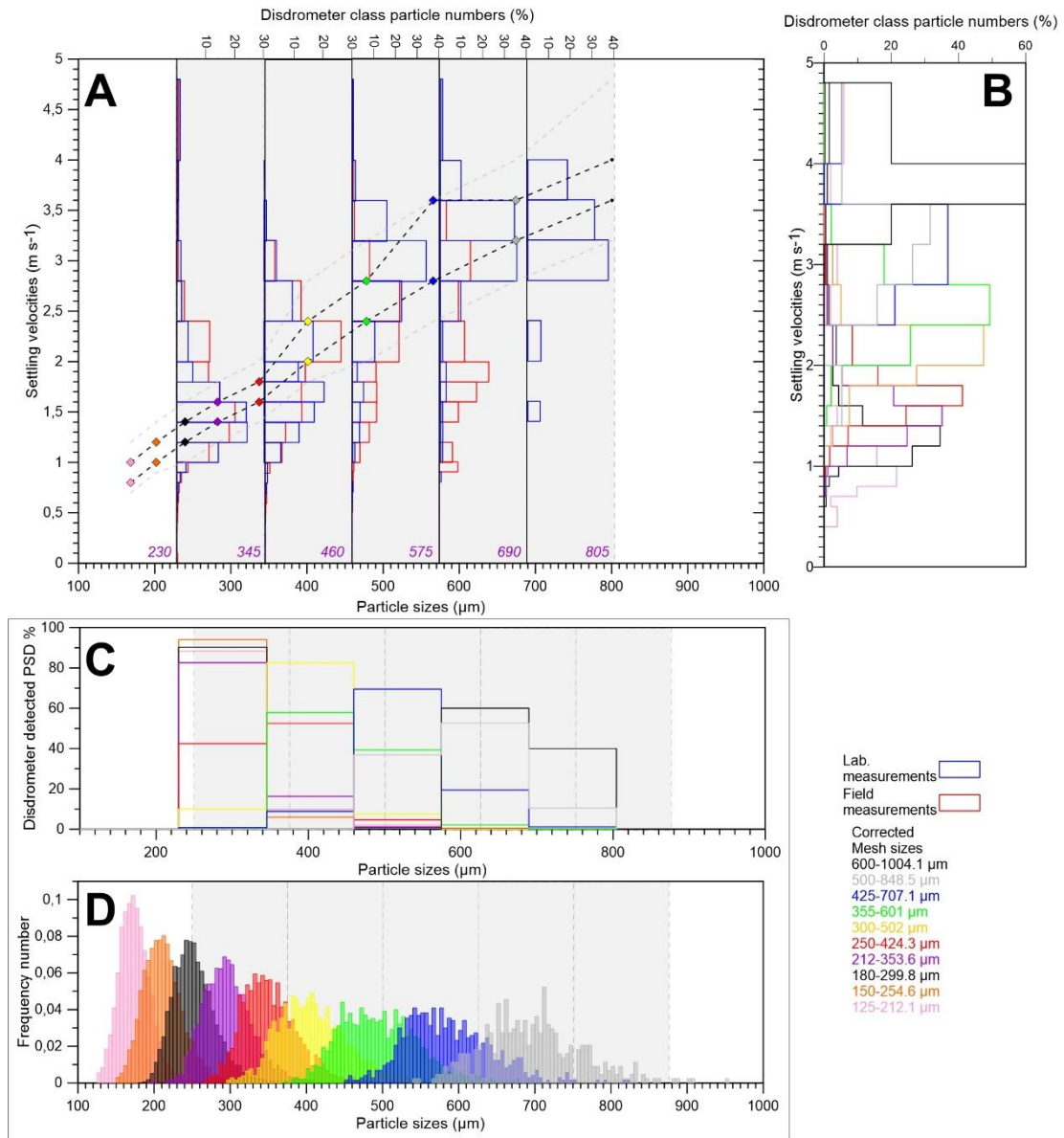
474

475 **Figure 9:** Settling velocity as a function of particle size classes measured by disdrometer during two
 476 ash fallout events at Stromboli. **A)** at 12:46 UTC (10/02/2015) and **B)** at 16:01 (10/03/2015). The color
 477 code represents the sum of the detected number of particles inside each class of velocities (y axis) and
 478 sizes in circle-equivalent diameter (D_{CE} , x axis).

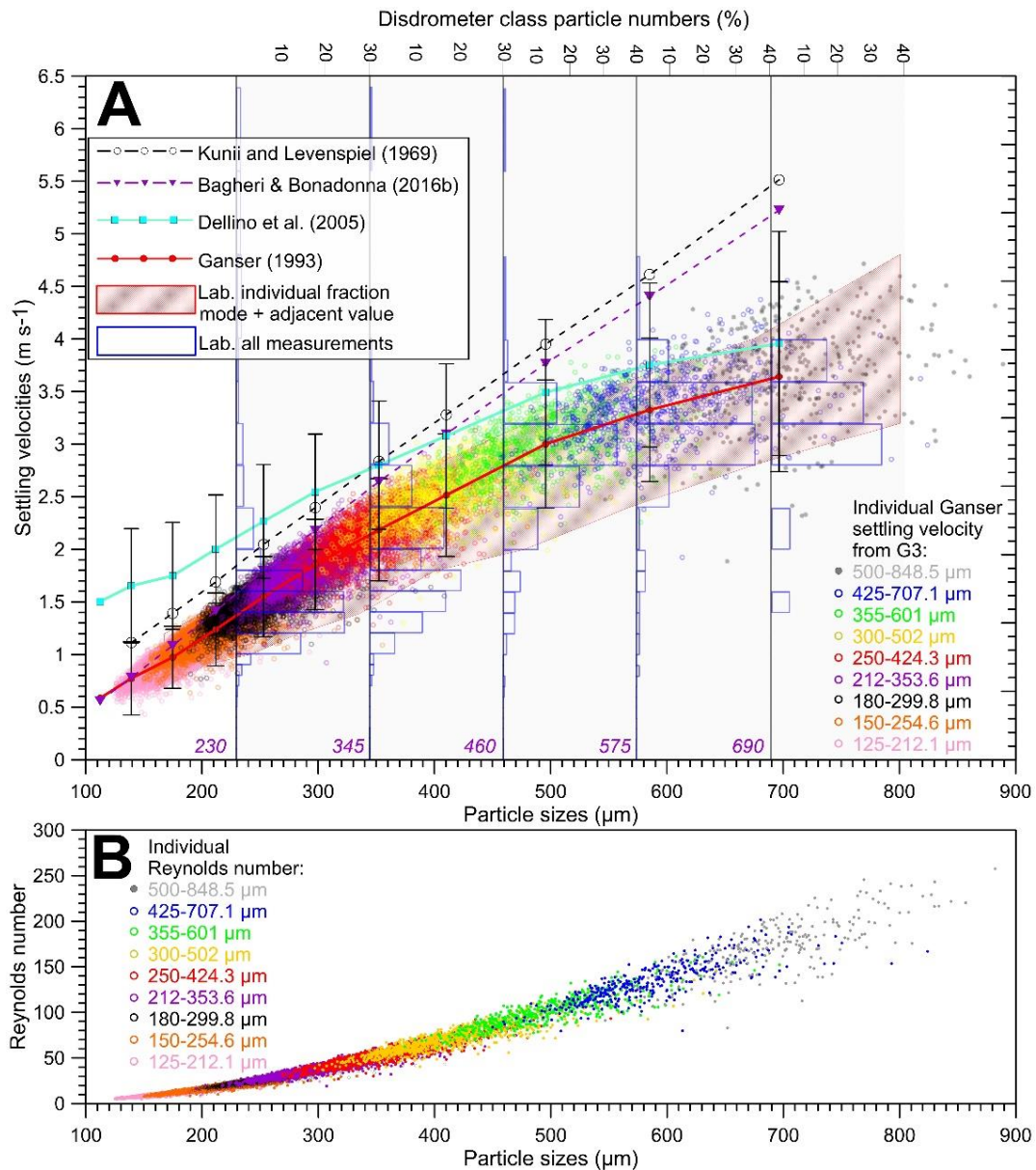
479 **4.2 Laboratory experiments on ash settling velocity**

480 V_T of dropped individual ash particles from the different sieved fractions is measured by
 481 the disdrometer under laboratory conditions of no wind. As expected, V_T distributions for each
 482 sieved fraction (**Figure 10A** and **10B**) are unimodal. The most frequently measured V_T increases
 483 with increasing D_{CE} from $0.95 \text{ m s}^{-1} \pm 0.05 \text{ m s}^{-1}$ for 125-212 μm , to $3.8 \text{ m s}^{-1} \pm 0.1 \text{ m s}^{-1}$ for
 484 600-1000 μm (Black dashed line in **Figure 10A**). Nevertheless, the spread of V_T above and
 485 under the modal V_T values in each size class (grey dashed line in **Figure 10A**) highlights the
 486 aforementioned heterogeneity of PSDs and particle shapes shown by **Figures 3** and **Figure 7**,
 487 respectively, in each sieved fraction (**Figure 10D**). Moreover, the individual detected PSDs
 488 from the disdrometer are in broad agreement with the G3 PSDs, taking into account the ratio
 489 between D_{CE} and $(L+I)/2$ (**Figure 10C** and **10D**). Likewise, a comparison of the mode and
 490 adjacent values of V_T of each sieved fraction (**Figure 10B** and dashed lines in **Figure 10A**), or

491 all measurements of V_T , shows broad agreement between values recorded in control experiments
 492 and in the field (blue and red histograms in **Figure 10A**, respectively). This highlights, in turn,
 493 the quality of the disdrometer data and the broad agreement between field and laboratory
 494 measurements. However, the distribution of field V_T of the 575-690 μm class appears to be
 495 bimodal: modal V_T measured in the lab matches the field mode at 3 m s^{-1} while most of the
 496 coarse particles in the field fell at lower V_T (mode at $V_T = 1.9 \text{ m s}^{-1}$).



497
 498 **Figure 10: A)** Settling velocities measured by disdrometer in laboratory conditions (blue histograms)
 499 and in the field (red histograms) for every sieved fraction from the 1601_summit sample. Dashed lines
 500 encompass the most frequently measured velocities (mode, bold black line) and adjacent classes (grey
 501 line). **B)** Histogram of settling velocities recorded by the disdrometer in each sieve class. **C)** Detected
 502 PSD (in percentage) and **D)** G3-derived PSDs (in frequency) of each sieve fraction.



503

504 **Figure 11:** A) Average settling velocities measured by disdrometer in laboratory conditions (red area
 505 encompassing mode and adjacent velocity values, blue histograms for all measured velocities) and
 506 calculated with empirical models (curves) using the morphological parameters' average values
 507 obtained from the G3 optical analyses. Best match of the Ganser (1993) model (red curve) with the
 508 disdrometer data. V_T calculated with the Ganser (1993) model for all analyzed particles of the
 509 1601_summit sample in each sieve fraction are displayed with colored dots. Error bars correspond to
 510 the standard error of the mean for every size class of particle V_T . B) Individual Reynolds number
 511 calculated with the Ganser (1993) drag equation as a function of all analyzed particle sizes of the
 512 1601_summit sample.

513 4.3 Empirical modeling

514 We compare 1601_summit ash V_T measured under laboratory conditions against the four
 515 empirical models described in section 2 (Figure 11A). Using the average ϕ , D_{CE} values and

516 densities found for each sieved fraction (**Appendix C**), we find that the Ganser model best
517 describes the increase in V_T for particles with D_{CE} from 125 to more than 800 μm in our data.
518 As shown in the preceding sections, the heterogeneity of particle shapes, sizes and densities is
519 the cause of the spread of settling velocity measurements either in the field or under laboratory
520 conditions. Therefore, we used the G3-inferred individual particle shape parameters to initialize
521 the Ganser model.

522 **5. Discussion**

523 **5.1 Empirical model validation**

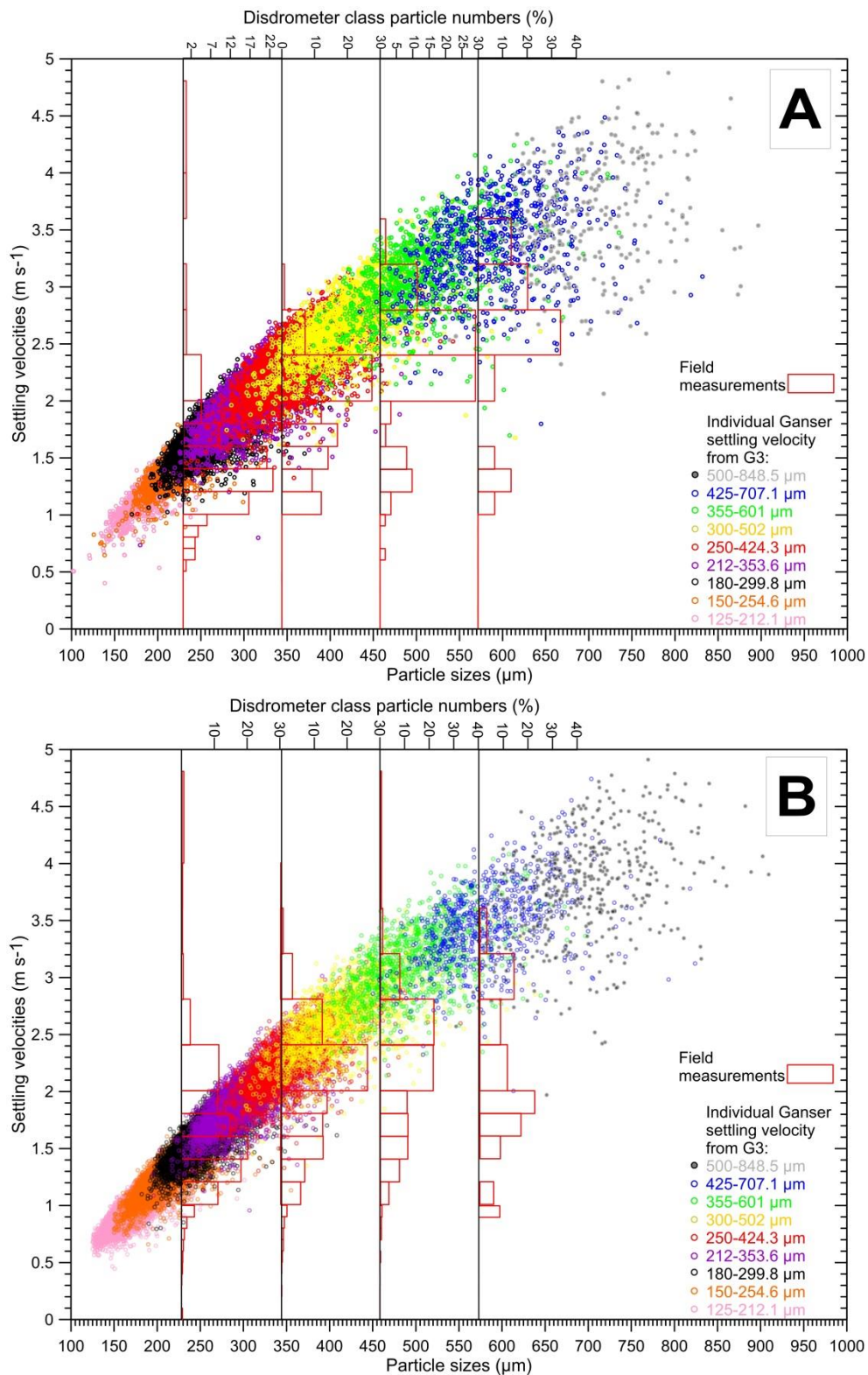
524 The combination of empirical models describing V_T of non-spherical particles permits
525 identification of the effects of physical ash particle characteristics such as size, density and
526 shape on the V_T calculation and also highlights the limits and strengths of each model. As
527 described in Beckett *et al.* (2015), V_T empirical models are mainly sensitive to ash PSD, whereas
528 their sensitivity to the shape and density is of lesser importance, but still relevant for precise V_T
529 modeling. Knowing the PSDs of ash fallout samples at high size-resolution allows
530 quantification of the sensitivity of such models to particle shape and density with a higher
531 precision. For the models of Ganser (1993) and Bagheri & Bonadonna (2016b), the main
532 parameter controlling V_T is the shape parameter used to calculate the drag coefficient. Ganser's
533 model requires the particle sphericity ϕ , whereas the Bagheri & Bonadonna model requires 3-
534 D particle measurements such as lengths of L , I and S axes. Because the short axis (S) is not
535 measured by the G3 optical analysis in 2-D, we had to hypothesize S as equal to the intermediate
536 (I) axis. This assumption tends to overestimate V_T in the Bagheri & Bonadonna model.
537 Nevertheless, in order to obtain similar V_T values between both models, S must be between $0.4I$
538 and $0.1I$. Such S/I ratios, no matter the L values, would correspond to thin or tabular particle
539 shapes, which do not characterize the average shape of our analyzed dense ash particles. Hence,
540 the methodology and analyzed particles used in this study do not permit us to use the Bagheri
541 & Bonadonna model for modeling terminal settling velocities.

542 There are two explanations for the better agreement between our measurements and the
543 velocities of Ganser (1993). First, regarding the abundant presence of dense ash fragments with
544 regular and rounded shapes (**Figures 7A, 7B, 7C and 7D**), the sphericity ϕ of Riley *et al.* (2003)

545 appears to be the optimal parameter to describe our grain population among the 6 ash samples.
546 Such a parameter is known to be well suited for the accuracy of Ganser's V_T equation (Alfano
547 *et al.*, 2011).

548 Secondly, values of V_T calculated from empirical models depend on the accuracy of the
549 shape factors used to determine the drag coefficient. φ is calculated from the particle area (*i.e.*
550 linked to its shape) but also its perimeter, which strongly depends on the small scale particle
551 roughness. For example, in Dioguardi *et al.* (2017), a 3-D sphericity is defined using X-ray
552 microtomography. Their sphericity values are much lower ($\varphi < 0.434$) than those obtained by
553 2-D analyses owing to the high spatial resolution that takes into account the particle roughness
554 at a very small scale. The G3 is less precise than X-ray microtomography for measuring small
555 scale particle asperities, indicating that the variations of φ are mainly due to changes in particle
556 shapes rather than in their roughness (Dioguardi *et al.*, 2018). Moreover, Strombolian ash
557 particles have small-scale roughness as in the study of Ganser (1993). Taken together, these
558 observations explain why, using our methodology, the best model describing V_T , measured by
559 the disdrometer over the largest interval of ash sizes, is the Ganser model. Using the
560 morphological parameters from our G3 optical analyses, **Equation 14** in Dellino *et al.* (2005)
561 is thus valid for coarse ash and lapilli, which remain sparse at Stromboli. Indeed, **Equation 14**
562 is established for a set of particles having a Reynolds Number > 60 -100 (Dioguardi *et al.*, 2018),
563 a range which corresponds to 5-10% of ash particles among the 1601_summit sample with D_{CE}
564 larger than 360 to 560 μm (**Figure 11B**).

565 V_T calculated with the Ganser model for every analyzed particle for the 1246_roc and
566 1601_summit samples is in good agreement with ash V_T measured in the field (**Figures 12A**
567 and **12B**) and under laboratory conditions. However, as observed in **Figures 9** and **10**, small V_T
568 are also observed in the upper disdrometer classes above 460 μm in both contexts. Those V_T
569 can be due to several effects: (i) the V_T being calculated from the crossing times of particles.
570 Ash particles might not have fallen perpendicularly to the laser sheet, *i.e.* non-vertical
571 trajectories, possibly due to the wind, causing longer crossing periods and thus lower V_T . (ii)
572 As shown by Bagheri & Bonadonna (2016b), particles may fall with their longest axis
573 perpendicular to their settling axis but may also oscillate and rotate according to this axis
574 resulting in varying crossing times corresponding to one of the 3-D axes of the particles. It is
575 unclear why any of these processes would have affected mainly coarser particles.



576

577 **Figure 12:** Individual particle V_T calculated with the model of Ganser (1993) based on sphericities (ϕ)
 578 and particle sizes measured by G3 in each sieved fraction (color code) for the October 2 2015 at 12:46
 579 UTC (A) and October 3 2015 at 16:01 UTC (B) fallout events. Associated ash deposits, 1246_roc and
 580 1601_summit samples respectively, were collected from ground tarps immediately after each fallout
 581 event. The distribution of disdrometer velocities measured in the field is shown in histograms for
 582 comparison.

583 5.2 Application of particle shape and disdrometer measurements to radar retrievals

584 During our measurement campaign, a 3 millimeter-wave Doppler radar was used in
585 addition to the disdrometer to record ash plumes dynamics and quantify ash concentrations
586 (Donnadieu *et al.*, 2016). Inside a radar beam, when a continuously emitted electromagnetic
587 wave encounters ash particles, its backscatter towards the radar induces a signal, the power of
588 which is used to calculate a reflectivity factor Z . By assuming that the target PSD in the probed
589 radar volume is composed of homogeneously distributed spherical ash particles with known
590 diameter D (Sauvageot, 1992):

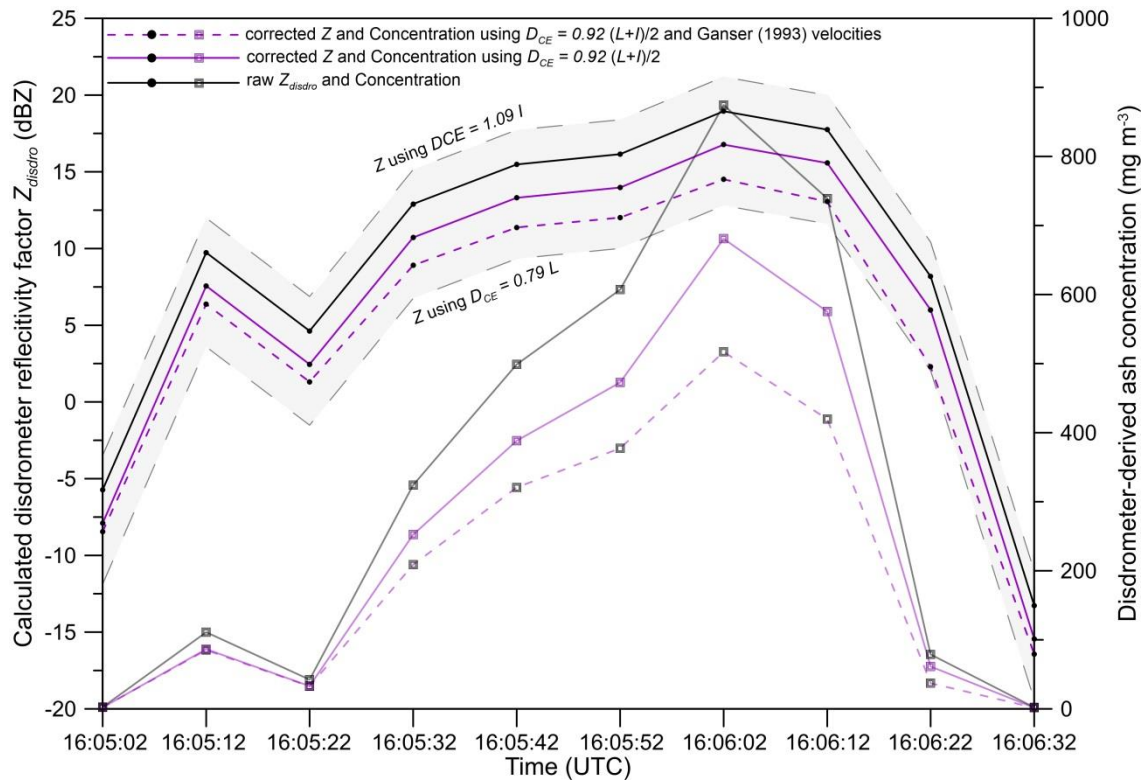
$$591 \quad Z = \int_{D_{\min}}^{D_{\max}} N(D)D^6 dD.$$

592 (15)

593 Z characterizes the volcanic mixture remotely probed by the radar beam and directly
594 reflects the particle volume concentration, however the strong contribution of spherical particle
595 sizes (D^6) and lesser contribution of particle amounts ($N(D)$) of each size cannot be isolated
596 without further constraints. One potential application of accurately characterizing volcanic
597 particle sizes is to refine radar retrievals. Disdrometer measurements of the number of
598 individually detected particles, their V_T and sizes allows estimation of radar reflectivity factors
599 and associated ash concentrations. Thus, the coupling of radar and optical disdrometer methods,
600 as implemented in meteorology (Marzano *et al.*, 2004; Maki *et al.*, 2005), will refine ash mass
601 load retrievals from radar remote sensing of ash plumes and their fallout.

602 The methodology applied in this study to characterize volcanic particle shapes improves
603 the interpretation of disdrometer outputs for a more accurate radar reflectivity estimation. The
604 combination of disdrometer measurements of $v_i = V_T$ and number of detected particles is used
605 to infer a particle number density per unit volume $N(D)$ (**Equation 4**), which is used, in turn,
606 to automatically calculate Z from the measured sizes of particles detected by the disdrometer.
607 Under the assumption that particles fall with their L and I axes in the beam plane (i.e.
608 horizontal), the raw disdrometer reflectivity, Z_{disdro} , is calculated directly from the detected
609 diameter, therefore assuming spherical particles, so that Z_{disdro} is biased for non-spherical
610 particles depending on their orientation when crossing the beam. For example, the size of
611 particles crossing the beam with their longest axis (L) normal to the detectors alignment are
612 overestimated, and so is Z . Contrastingly, Z is underestimated when the intermediary axis is

613 seen by the beam. From our morphological study, carried out statistically on a large number of
 614 ash particles, we conclude that the conversion of $D_{CE} = 0.92 (L+I)/2$ is appropriate (R^2 of 0.999
 615 in **Figure 7C**) and can be used to constrain disdrometer reflectivities of Strombolian ash.



616
 617 **Figure 13:** Disdrometer reflectivity factor (Z_{disdro} , dot lines) and ash concentration (line with squares)
 618 calculated from disdrometer measurements as a function of time during an ash fallout event on October
 619 3 2015 (event 16:01 UTC corresponding to the sample 1601_summit). The black dot line corresponds
 620 to the raw Z_{disdro} calculated with no particle shape conversion, whereas the grey area represents the raw
 621 reflectivity if the disdrometer detected all particles respectively along their longest and intermediate
 622 axis. Purple line and purple dashed lines indicate Z and concentration values by correcting,
 623 respectively, only particle sizes in circle-equivalent diameter (D_{CE} inferred from G3 analyses), or both
 624 D_{CE} and settling velocities using the Ganser model (1993).

625 Comparing Z_{disdro} with and without (i) conversions for circle-equivalent diameter and
 626 (ii) model-derived V_T (**Figure 13**) shows the respective influence of these two parameters (that
 627 are used to calculate $N_i(D_i)$ in **Equation 4** and **Equation 15**) on the reflectivities. Correcting
 628 the PSD by using the ratio $(L+I)/2$ shows a decrease of 2.18 dBZ in average (**Figure 13**). This
 629 highlights the necessity to physically characterize non-spherical particles, such as volcanic ash,
 630 and then to correct disdrometer data accordingly when comparing with reflectivities measured
 631 *in situ* inside the ash mixtures (*i.e.* plume and fallout).

632 Furthermore, the best fitting Ganser (1993) model V_T measured by the disdrometer in
 633 the field can be used to calculate V_T and correct for the data scattering, in particular the outlying

634 low V_T of coarse ash measured in the field (**Figures 9** and **10**). This results in a further average
635 difference of 1.8 dBZ compared to the PSD correction using the conversion of $(L+I)/2$, *i.e.* a
636 total decrease of 4 dBZ using PSD and V_T correction with respect to reflectivities calculated
637 from raw data.

638 Finally, detected ash concentration C_{ash} may be calculated using the following equation:

$$639 \quad C_{ash} = \frac{\pi \rho_i}{6} \int_{D_{i\min}}^{D_{i\max}} N_i(D_i) D_i^3 dD_i . \quad (16)$$

640 As a result, disdrometer-derived ash concentrations span between 2.23 and 874.52 mg
641 m^{-3} without any correction for diameters (**Figure 13**). Using the $(L+I)/2$ conversion leads to
642 smaller ash concentrations to 1.73-680.79 mg m^{-3} (average difference of $22.155 \pm 0.003\%$).
643 Moreover, the use of Ganser's equation (1993) decreases conversion-derived and initial
644 concentrations of 14.32% and 33.3%.

645 Despite the main dependence of ash reflectivity factors and concentrations on particle
646 size (D^6 in **Equation 15** and D^3 in **Equation 16**), low velocities measured by the disdrometer
647 in the field seem to have a non-negligible impact on the quantitative retrievals obtained from
648 disdrometer retrievals.

649 Thus, as in meteorology, considering similar PSDs between the atmospheric volumes
650 possibly probed by the radar and fallout measurements at ground level (Marzano *et al.*, 2004;
651 Maki *et al.*, 2005), the disdrometer-inferred reflectivity factors provide a reasonable first-order
652 quantification of ash concentrations inside volcanic ash plumes. The next step is to compare
653 disdrometer-inferred reflectivity factors with reflectivity factors measured by radar inside the
654 ash plumes and then estimate the spatial distribution of the ash mass load, one of the most
655 crucial source term parameters.

656 **5.3 Validation and limitation of disdrometer data**

657 Despite a disdrometer lower detection limit of 230 μm in D_{CE} (*i.e.* only coarse ash is
658 detected), the morpho-grainsizer G3 measurements and disdrometer measurements yield
659 similar PSD modes. The low proportion of coarse ash larger than 690 μm detected by the
660 disdrometer can be explained by the difference of spatial resolution between the instrument and
661 the tarp used to sample the fallout (*i.e.* a laser sheet surface of 0.0054 m^2 compared to a 0.4 m^2

662 tarp). Indeed, the relatively low percentage of particles coarser than 575 μm (between 0.21 and
663 13.75% in **Figure 5**) can lead to an under-sampling of such sparse particles by the disdrometer.

664 The tests under laboratory conditions using empirical models validate the disdrometer
665 V_T measurements in the field. However, field measurements show a high dispersion of V_T . With
666 a modal value of $2.2 \pm 0.2 \text{ m s}^{-1}$, measured V_T in the class 345-460 μm (**Figure 10**) range
667 between $1 \pm 0.1 \text{ m s}^{-1}$ and $3 \pm 0.2 \text{ m s}^{-1}$. Such a high variability is also seen with no-wind
668 conditions in the laboratory (**Figures 10** and **11**). We attribute such variations to fluctuations in
669 vertical wind and beam crossing trajectories, but also due to the high variability of particle
670 shapes, sizes, and densities that lead to many possible combinations of interactions with the
671 ambient fluid that can induce 3-D changes in particle rotation (Bagheri & Bonadonna, 2016b).

672 **6. Conclusive remarks**

673 This study presents an exhaustive characterization of the ash produced during
674 explosions at Stromboli. We also perform an inter-comparison of empirical settling velocity
675 models to validate terminal settling velocities measured by an optical disdrometer in the field.

676 The use of mechanical sieving and the morpho-grainsizer MORPHOLOGI G3 reveals
677 the need to consider the geometrical influence of the sieve meshes in the case of non-spherical
678 and rough ash particles. We propose the use of the lower mesh size and the diagonal of the
679 upper mesh size of the mechanical sieve to constrain each PSD fraction.

680 We further propose a method to obtain a total PSD by combining the morpho-grainsizer
681 high-resolution PSD with the weight percentages measured by the mechanical sieving. Our
682 analyses reveal dense and homogenous ash particles in terms of textural and morphological
683 roughness for a set of type 2 eruptions from proximal to medial distance (2 km) from the summit
684 vents of Stromboli. The non-spherical ash particles sampled at Stromboli have their circle-
685 equivalent diameter equal to 0.79 times their longest axis, 1.09 times their intermediate axis on
686 average and 0.92 times their average 2-D dimension $(L+I)/2$. Moreover, we show that particle
687 sphericity tends to slightly decrease with increasing D_{CE} . This observation may have strong
688 implications for ash dispersion modeling, even in the case of moderate-sized eruptions. Indeed,
689 the residence time of airborne ash particles has been shown to be highly dependent on their
690 shapes, mostly for coarse ash with $D_{CE} > 200 \mu\text{m}$ (Beckett *et al.*, 2015; Saxby *et al.*, 2018).

691 Disdrometer settling velocity measurements of sieved ash fractions under laboratory
692 conditions are in broad agreement with the field settling velocities. This validates the
693 volcanological application of the meteorology-designed OTT Parsivel² disdrometer for the
694 detection and real-time recording of ash particle fallout size and settling velocities. Its
695 limitations mainly concern the lower threshold in size measurements (230 microns in circle-
696 equivalent diameter at Stromboli) and the width of each size class (*i.e.* around the 115 μm size
697 class when used for ash measurements).

698 Empirical models used to calculate terminal settling velocities are highly dependent on
699 the input shape parameters and their accuracy. Whereas the Bagheri & Bonadonna (2016b) and
700 Dioguardi *et al.* (2017) models are based on high precision 3-D analyses of particles, the lower-
701 resolved G3-derived sphericities from fast routine 2-D optical analyses are better matched to
702 the Ganser model (1993). This is due to the fact that the output of our methodology more
703 accurately reproduces the parameters used in Ganser (1993) to model terminal settling
704 velocities.

705 Finally, the empirical results obtained with the Ganser model (1993) provide the best fit
706 with the terminal settling velocities measured by the disdrometer and highlight the capability
707 of such an instrument to operationally monitor volcanic ash sizes and their settling velocities
708 with higher time resolution (10 s) than other in-situ methods. Despite underestimating coarse
709 ash settling velocities, possibly due to particle interactions with the ambient fluid, our physical
710 ash characterization emphasizes the need to constrain the size of both intermediate and long
711 particle axes (I and L) and their settling velocities in order to calculate radar reflectivity factors.
712 This underlines the important role of disdrometers in the field to constrain radar data for ash
713 plume monitoring and mass load retrieval.

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726

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870 **Appendix A: Mass (g) and mass percentage obtained for each sieved fraction for the six ash**
871 **samples.**

1/2 Φ Diameter (µm)	1601_summit		1246_roc		1530PL		1550_summit		1636_summit		1042-1252_roc		
	Mass (g)	wt%	Mass (g)	wt%	Mass (g)	wt%	Mass (g)	wt%	Mass (g)	wt%	Mass (g)	wt%	
2000-2800											0,0299	0,4396	
1400-2000	0,0099	0,0392									0,0426	0,6264	
1000-1400	0,0055	0,0218	0,0064	0,1287							0,0111	0,1632	
710-1000	0,0185	0,0733	0,0050	0,1006			0,0004	0,5891			0,0406	0,5970	
500-710	0,6204	2,4590	0,1288	2,5909	0,0003	0,1157	0,0006	0,8837	0,0040	0,6416	0,0318	0,4676	
355-500	4,4479	17,6295	1,2990	26,1305	0,0010	0,3855	0,0012	1,7673	0,0280	4,4915	0,0391	0,5749	
250-355	7,9853	31,6503	2,3787	47,8496	0,0087	3,3539	0,0033	4,8601	0,1441	23,1152	0,2829	4,1597	
180-250	5,5236	21,8932	0,7910	15,9117	0,0606	23,3616	0,0120	17,6730	0,2052	32,9163	1,2834	18,8710	
125-180	3,6909	14,6291	0,1902	3,8260	0,1128	43,4850	0,0372	54,7865	0,1961	31,4565	2,1109	31,0385	
90-125	1,6927	6,7091	0,0677	1,3618	0,0482	18,5813	0,0124	18,2622	0,0382	6,1277	1,4896	21,9030	
63-90	0,7670	3,0401	0,0431	0,8670	0,0227	8,7510	0,0008	1,1782	0,0062	0,9945	0,8614	12,6660	
0-63	0,4681	1,8553	0,0613	1,2331	0,0051	1,9661			0,0016	0,2567	0,5776	8,4930	
1/4 Φ Diameter (µm)	1601_summit		1246_roc		1530PL		1550_summit		1636_summit		1042-1252_roc		
Mass (g)	wt%	Mass (g)	wt%	Mass (g)	wt%	Mass (g)	wt%	Mass (g)	wt%	Mass (g)	wt%	Mass (g)	wt%
2360-2800											0,0298	0,4501	
2000-2360											0,0000	0,0000	
1600-2000											0,0328	0,4954	
1400-1600											0,0108	0,1631	
1180-1400											0,0058	0,0876	
1000-1180											0,0060	0,0906	
850-1000											0,0209	0,3156	
710-850							0,0004	0,7156			0,0171	0,2583	
600-710	0,0726	0,3064	0,0173	0,3660	0,0000	0,0000	0,0000	0,0000	0,0016	0,2638	0,0090	0,1359	
500-600	0,5189	2,1897	0,1034	2,1874	0,0001	0,0400	0,0003	0,5367	0,0022	0,3627	0,0171	0,2583	
425-500	1,7265	7,2857	0,4670	9,8794	0,0004	0,1601	0,0004	0,7156	0,0052	0,8572	0,0168	0,2537	
355-425	2,6035	10,9865	0,7508	15,8832	0,0003	0,1200	0,0003	0,5367	0,0240	3,9565	0,0242	0,3655	
300-355	3,7705	15,9112	1,6416	34,7281	0,0027	1,0804	0,0013	2,3256	0,0699	11,5232	0,0865	1,3064	
250-300	3,2857	13,8654	0,5719	12,0986	0,0047	1,8808	0,0016	2,8623	0,0537	8,8526	0,1236	1,8667	
212-250	3,8763	16,3576	0,6160	13,0315	0,0213	8,5234	0,0043	7,6923	0,1360	22,4200	0,5249	7,9274	
180-212	2,3582	9,9514	0,2050	4,3368	0,0414	16,5666	0,0081	14,4902	0,0893	14,7214	0,5555	8,3896	
150-180	2,3027	9,7172	0,1390	2,9405	0,0703	28,1313	0,0159	28,4436	0,1316	21,6947	1,1451	17,2942	
125-150	1,3902	5,8665	0,0480	1,0154	0,0416	16,6467	0,0132	23,6136	0,0571	9,4131	0,9691	14,6361	
112-125	0,8370	3,5321	0,0238	0,5035	0,0198	7,9232	0,0040	7,1556	0,0186	3,0663	0,6154	9,2942	
90-112	0,9551	4,0304	0,0286	0,6050	0,0234	9,3637	0,0051	9,1234	0,0117	1,9288	0,8304	12,5413	
75-90					0,0122	4,8820	0,0008	1,4311	0,0038	0,6264	0,5278	7,9712	
63-75					0,0075	3,0012	0,0001	0,1789	0,0011	0,1813	0,3712	5,6061	
0-63							0,0001	0,1789	0,0008	0,1319	0,6815	10,2925	

873 **Appendix B:** Disdrometer classification of settling velocities.

Class index	Settling velocity (m s ⁻¹)	Class spreading (m s ⁻¹)
1	0.05	0.1
2	0.15	0.1
3	0.25	0.1
4	0.35	0.1
5	0.45	0.1
6	0.55	0.1
7	0.65	0.1
8	0.75	0.1
9	0.85	0.1
10	0.95	0.1
11	1.1	0.2
12	1.3	0.2
13	1.5	0.2
14	1.7	0.2
15	1.9	0.2
16	2.2	0.4
17	2.6	0.4
18	3	0.4
19	3.4	0.4
20	3.8	0.4
21	4.4	0.8
22	5.2	0.8

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876 **Appendix C:** Average circle equivalent diameters $\overline{D_{CE}}$, Sphericities $\overline{\phi}$ and densities $\overline{\rho}$ found

877 for the two summit samples: 1246_roc and 1601_summit.

1246_roc Corrected Sieved fraction (μm)	$\overline{D_{CE}}$ (μm)	Std. error	$\overline{\phi}$	Std. error	$\overline{\rho}$ (kg m^{-3})
500-600	698.97	± 3.77	0.706	± 0.005	-
425-500	588.36	± 2.3	0.745	± 0.003	2645
355-425	506.01	± 1.73	0.765	± 0.002	2710
300-355	415.56	± 1.24	0.770	± 0.002	2737
250-300	355.28	± 0.68	0.780	± 0.001	2708
212-250	307.82	± 0.58	0.776	± 0.001	2782
180-212	262.74	± 0.42	0.777	± 0.001	2811
150-180	228.19	± 0.75	0.765	± 0.002	2811
125-150	185.86	± 0.80	0.763	± 0.003	2811
1601_summit Corrected Sieved fraction (μm)	$\overline{D_{CE}}$	Std. error	$\overline{\phi}$	Std. error	$\overline{\rho}$ (kg m^{-3})
500-600	696.59	± 3.06	0.737	± 0.003	-
425-500	585.16	± 2.08	0.764	± 0.002	2683
355-425	495.67	± 1.59	0.777	± 0.002	2728
300-355	410.08	± 1.13	0.765	± 0.002	2743
250-300	352.31	± 0.73	0.759	± 0.001	2772
212-250	297.37	± 0.51	0.759	± 0.001	2774
180-212	252.94	± 0.42	0.744	± 0.001	2792
150-180	211.87	± 0.35	0.730	± 0.001	2737
125-150	174.65	± 0.23	0.714	± 0.001	2724

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